Evaluating the influence of CCA and DOT wood preservative treatments on one component laminated *P. patula* polyurethane bond performance

by

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Declaration

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Summary

With the increasing movement towards environmentally friendly and sustainable building materials in the construction industry, engineered wood products have become a preferred structural material. However, the adoption of engineered wood products as a building material could be curtailed by failure to identity and eliminate threats, such as biodeterioration agents (e.g. fungus, insects). Wood preservation offers the opportunity to improve wood resistance and increase product service, through the impregnation of wood preservative chemicals into the wood cell lumen and walls. Some engineered wood products are too large to be treated after manufacturing, which necessitates treatment before adhesive bonding of laminates. In other cases treatment before adhesion is preferable from a process and chemical retention perspective. However, the metallic and inorganic salt deposits of the preservatives may present complications, as they can interfere with bond formation between the wood substrates and adhesive. The interference of these deposits may lead to poor bond strength and durability and could lead to product failure and not meeting the standard requirements.

This research involved evaluating popular wood preservation treatments on the bond line performance of *Pinus patula* wood bonded with a one-component polyurethane (PUR) adhesive. The specific objectives of the experimental study were as follows:

- Evaluate the effects of chromium copper arsenic (CCA) and disodium octaborate tetrahydrate (DOT) wood preservatives and *Pinus patula* wood properties (density, sapwood/heartwood) on the bond performance of one component polyurethane bonded laminates;
- Evaluate the influence of wood properties of *Pinus patula* (sapwood, heartwood, and density) on retention rate.

The experimental design consisted of four factors (treatment chemical, concentration, wood density and sapwood/heartwood ratio). The treatment chemicals had different treatment levels: CCA (2% and 4% concentration), DOT (1.67% and 3.30% concentration) and an untreated control. The density had two levels - lower than 462 kg/m^3 and higher than 473 kg/m^3 and the wood type was separated into two levels, sapwood only specimens and specimens with more than 35% heartwood. In total, the experimental study had 20 groups which were tested for bond shear strength and delamination.

To realise the objectives of the experimental research, the wood impregnation process was adopted from SANS 10005 (2016), whilst the material specifications and laminate manufacturing procedures were adopted from SANS 10183-4-2 (2009) and ASTM D905 (2008). The performance of the PUR adhesive bonds was evaluated and measured through standardised test methods including shear strength, wood failure percentage and resistance to delamination by accelerated exposure.

All groups in the experiment met the average requirements of EN 14080 (2013), both for shear strength and wood failure percentage. Interestingly, the 4% CCA treated specimens displayed superior shear strength in comparison to the control and 2% CCA specimens. However, with increasing concentration levels of CCA, the wood failure percentage was negatively affected. Overall, the DOT-treated specimens displayed more consistent performance in comparison to CCA specimens, in terms of shear strength and wood failure. The results also indicated that wood properties play a significant role in the strength of bonds. High-density samples

produced higher shear strength. However, one of the unexpected findings was that in most cases heartwood specimens showed a higher shear strength (whether treated with CCA or DOT or untreated) in comparison to sapwood.

In terms of bond durability, all the treated and untreated (control) test blocks met the requirements of EN 14080 (2013), as the average total delamination did not exceed 10% in length (mm) in any of the groups. Overall, the DOT treated samples were found to have a better resistance to delamination in comparison to CCA treated samples. The results also indicated that with increasing concentration levels of CCA, delamination increased.

With regards to the effect of wood properties on retention rate, the results showed that sapwood had a higher retention capacity than heartwood for both preservatives (CCA and DOT). Density was also found to have a significant effect on retention with the retention rate being much lower in most high-density wood samples when compared to low density-wood samples. Such findings highlight the importance of understanding the treatability behaviour/response of various parts of wood (e.g. sapwood, heartwood), anatomical characteristics (e.g. thick or thin cells walls) and size, in order to ensure the required or targeted retention and penetration is achieved during treatment.

Overall, the shear strength, wood failure and delamination results suggested that engineered wood products can be produced from CCA and DOT treated *Pinus patula*. However, the concentration levels should be carefully selected, as the study found that with increasing concentration levels, delamination also increased.

Keywords: *Pinus patula*, CCA, DOT, retention rate, engineered wood products, shear strength, delamination, wood failure percentage, sapwood, heartwood, density, 1C-PUR

Opsomming

Met die toenemende beweging na omgewingsvriendelike en volhoubare boumateriaal in die konstruksiebedryf het saamgestelde houtprodukte 'n voorkeurmateriaal geword. Die aanvaarding van houtprodukte as 'n boumateriaal kan egter beperk word deur bedreigings soos degradasie-agente insluitende swamme en insekte. Houtpreservering bied die geleentheid om weerstand teen degradasie te verbeter en die diens van die produk te verhoog deur chemikalieë wat houtbeskermingsmiddels bevat in die lumen en selwande te deponeer. Sommige vervaardigde houtprodukte is te groot om na vervaardiging behandel te word, wat beteken dat behandeling voor die adhesieproses moet plaasvind. In ander gevalle is behandeling voor adhesie verkieslik vanuit 'n proses- en chemiese retensieperspektief. Die metaal- en anorganiese soutafsettings van die preserveermiddels kan egter komplikasies oplewer aangesien dit die vorming van bindings tussen die houtvesels en kleefmiddel kan belemmer. Die inmenging van hierdie afsettings kan lei tot swak bindingssterkte en duursaamheid en kan lei tot produkte wat nie aan die standaardvereistes voldoen nie.

Hierdie navorsing het die evaluering van gewilde houtbehandelings op die bindingskwaliteit van *Pinus patula*-hout wat met 'n een-komponent poli-uretaan (PUR) kleefmiddel gebind is, geëvalueer. Die spesifieke doelstellings van die eksperimentele studie was soos volg:

- Evalueer die effekte van chroomkoperarsenika (CCA) en dinatrium-oktaboraat tetrahidraat (DOT) houtpreserveermiddels en houteienskappe van *Pinus patula* (digtheid, spinthout / kernhout) op die bindingsprestasie van een-komponent-poli-uretaan laminate;
- Evalueer die invloed van houteienskappe van *Pinus patula* (spinthout / kernhout en digtheid) op die retensie van preserveermiddels.

Die eksperimentele ontwerp het bestaan uit vier faktore (behandelingschemikalie, konsentrasie, houtdigtheid en spintthout / kernhoutverhouding). Die behandelingschemikalieë het verskillende behandelingsvlakke gehad: CCA (2% en 4% konsentrasie), DOT (1,67% en 3,30% konsentrasie) en 'n onbehandelde kontrole. Die digtheid het twee vlakke gehad - laer as 462 kg/m^3 en hoër as 473 kg/m^3, en die houtsoort is in twee vlakke geskei, slegs spinthoute en monsters met meer as 35% kernhout. In totaal het die eksperimentele studie 20 groepe gehad wat getoets is vir die skuifsterkte en delaminasie van die binding.

Om die doelstellings van die eksperimentele navorsing te verwesenlik, is die houtimpregnasieproses vanaf SANS 10005 (2016) aangeneem, terwyl die materiaalpesifikasies en die vervaardigingsprosedures vir laminate vanaf SANS 10183-4-2 (2009) en ASTM D905 (2008) aangeneem is. Die werkverrigting van die PUR-kleefverbindings is geëvalueer en gemeet aan die hand van gestandaardiseerde toetsmetodes, insluitend skuifsterkte, persentasie houtbreek en weerstand teen delaminasie deur versnelde blootstelling.

Al die groepe in die eksperiment het aan die gemiddelde vereistes van EN 14080 (2013) voldoen, beide vir skuifsterkte en persentasie houtbreek. Interessant genoeg het die 4% CCA-behandelde monsters superieure skuifsterkte vertoon in vergelyking met die kontrole en 2% CCA-monsters. Met toenemende konsentrasievlakke van CCA, is die persentasie houtbreek egter negatief beïnvloed. Oor die algemeen het die DOT-behandelde monsters meer konsekwente prestasie getoon in vergelyking met CCA-monsters, wat die

skuifsterkte en houtbreek betref. Die resultate het ook aangedui dat houteienskappe 'n belangrike rol speel in die sterkte van bindings. Monsters met hoë digtheid het hoër skuifsterkte opgelewer. Een van die onverwagte bevindings was egter dat kernhoutmonsters in die meeste gevalle 'n hoër skuifsterkte vertoon (hetsy behandel met CCA of DOT of onbehandeld) in vergelyking met spinthout.

Wat die duursaamheid van die bindings betref, het al die behandelde en onbehandelde (kontrole) toetsblokke aan die vereistes van EN 14080 (2013) voldoen, aangesien die gemiddelde totale delaminasie in geen van die groepe meer as 10% was nie. Oor die algemeen is gevind dat die DOT-behandelde monsters 'n beter weerstand teen delaminering het in vergelyking met CCA-behandelde monsters. Die resultate het ook aangedui dat delaminering met toenemende konsentrasievlakke van CCA toegeneem het.

Wat die effek van houteienskappe op die retensietempo betref, het die resultate getoon dat spinthout 'n hoër retensievermoë as kernhout vir beide preserveermiddels (CCA en DOT) het. Daar is ook bevind dat digtheid 'n beduidende uitwerking op die retensie het, aangesien die retensietempo baie laer was in die meeste hoë-digtheid-houtmonsters, vergeleke met lae-digtheid-houtmonsters. Sulke bevindings beklemtoon die belangrikheid van die begrip van die behandelbaarheidsgedrag / reaksie van verskillende dele van hout (bv. spinthout, kernhout), anatomiese eienskappe (bv. dik of dun selwande) en grootte, ten einde te verseker dat die vereiste of doelgerigte behoud en penetrasie bereik word tydens behandeling.

Oor die algemeen het die skuifsterkte, houtbreek en delaminasie-resultate aangedui dat saamgestelde houtprodukte vervaardig kan word uit CCA en DOT-behandelde Pinus patula. Die konsentrasievlakke moet egter noukeurig gekies word, aangesien die studie bevind het dat delaminering met toenemende konsentrasievlakke ook verhoog het.

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Table of Contents

Declaration ..... **i**

Summary ..... **ii**

Opsomming ..... **iv**

Acknowledgements ..... **vi**

Table of Contents ..... **vii**

List of Figures ..... **x**

List of Tables ..... **xii**

List of abbreviations ..... **xiii**

Chapter 1 : Introduction **1**

1.1. Background to research question 1

1.2. Problem statement 2

1.3. Objectives 3

1.4. Brief Chapter Overview 3

1.5. Approach and procedure 3

1.6. Limitations and constraints of the experiment or study 3

Chapter 2 : Literature Review **4**

2.1. Engineered wood products 4

2.2. Wood preservation 5

2.2.1. Wood preservatives 5

2.2.2. Impregnation techniques 7

2.2.3. Absorption of preservatives in softwood 8

2.2.4. Effect of preservatives on chemical properties of wood 9

2.2.5. Effect of preservatives on mechanical properties of wood 10

2.2.6. Effect of preservatives on surface properties of wood 11

2.3. Wood adhesives 12

2.3.1. One component polyurethane adhesive 12

2.3.2. Penetration of adhesives 14

2.4. Factors influencing bond formation and performance 14

2.5. Wood-adhesive bond testing methods 19

2.5.1. Standards for wood-adhesive testing 19

2.5.2. Shear strength, resistance to delamination and wood failure percentage 21
2.6. Performance of glued treated wood 22
Chapter 3 : Materials and Methods 28
3.1. Materials 28
3.1.1. Wood 28
3.1.2. Adhesive 28
3.1.3. Wood preservative chemicals 29
3.2. Methods 29
3.2.1. Experimental design 29
3.2.2. Sample preparation 30
3.2.3. Heartwood/sapwood percentage determination 30
3.2.4. Density grouping/profiling 31
3.2.5. Wood impregnation process 31
3.2.6. Production of laminates 34
3.2.7. Test blocks 35
3.2.8. Performance test methods 37
3.2.9. Data analysis 42
Chapter 4 : Results and Discussions 43
4.1. Overview of shear strength, wood failure and delamination results 43
4.2. Relationship between wood properties and retention rate 47
4.3. Shear strength and WFP: effect of CCA and DOT preservatives 55
4.3.1. CCA and control shear strength 56
4.3.2. DOT and control shear strength 63
4.3.3. Comparison of treatments on shear strength and wood failure 67
4.4. Delamination: effect of CCA and DOT preservatives 71
4.4.1. Delamination of CCA and control blocks 71
4.4.2. Delamination of DOT and control blocks 74
4.4.3. Comparison of CCA and DOT treatments on delamination 74
Chapter 5 : Conclusion and Recommendations 77
5.1. Conclusions 77
5.2. Recommendations 78

Glossary **79**

Reference List **80**

APPENDICES **88**

APPENDIX A: Retention and shear strength results with detailed groups data..... 88

APPENDIX B: Retention and delamination results with detailed groups data 89

List of Figures

Figure 2-1: Common engineered wood products (Ramage et al., 2017). 4
Figure 2-2: Pressure treating cycles. 8
Figure 2-3: Softwood porosity structure (Milton, 1995). 9
Figure 2-4: Time-dependent contact angle with all the replicate data points for untreated and CCA-treated wood with distilled water used as a wetting liquid (Maldas and Kamdem, 1998). 12
Figure 2-5: Factors affecting bond strength and quality. 15
Figure 2-6: Bonding strength of Scotch pine wood according to the environment, sapwood-heartwood, and adhesive type (Kaygin and Tankut, 2008). 17
Figure 2-7: Shear strength results based on different moisture content levels (Gruver and Brown, 2006). .. 18
Figure 2-8: ASTM D905 (2003) test specimen configuration (d, e, f). 20
Figure 2-9: EN 392 (1995) test specimen configuration. 20
Figure 2-10: Bordered pit aperture showing a relative size of metal deposits to the opening through which adhesive flows (Vick, 1994). 23
Figure 2-11: Adhesion strength of untreated and treated samples (Ozdemir, Temiz and Aydin, 2015). 24
Figure 2-12: Mean block shear strength of CLT configurations by different adhesive types (bars with different letters are significant) (Lim, Tripathi and Tang, 2020). 25
Figure 2-13: Results of the average delamination for each wood treatment level/retention (Z=untreated, L= low 7.6 kg/m^3 and H= high 19.1 kg/m^3. 25
Figure 2-14: Effects of increased CCA retention on delamination of PRF bond lines (allowable maximum delamination = 5%) (Tascioglu, 2002). 26
Figure 3-1: Experimental procedure. 28
Figure 3-2: Heartwood and sapwood detection in specimens. 31
Figure 3-3: Modified empty cell treating cycle. 32
Figure 3-4: Delamination samples (left): A – 3.33% DOT, B – 1.67% DOT, C – 4% CCA, D – 2% CCA, E - Untreated; Shear samples (right): F – CCA treated and G – DOT treated samples. 34
Figure 3-5: Delamination laminates under pressure in a pneumatic press system. 35
Figure 3-6: Shear strength test blocks. 36
Figure 3-7: Delamination laminate (A) and 75mm test blocks (B). 36
Figure 3-8: Shear testing tool. 38
Figure 3-9: Wood failure evaluation of shear strength blocks. 39
Figure 3-10: Delamination test blocks in the drying oven. 40
Figure 3-11: Heartwood untreated (A) and sapwood CCA-treated (B) delamination test blocks (with marked delamination openings) after three impregnating-drying cycles. 41
Figure 4-1: Boxplot of mean shear strength values grouped by group number. 44
Figure 4-2: Mean plot of wood failure percentage values grouped by group number. 45
Figure 4-3: Mean plot of delamination values grouped by group number. The error bars indicate the variability/spread of the delamination data. 46
Figure 4-4: The interaction between wood type and density for retention rate (kg/m^3) of CCA and DOT preservative treated specimens. 50
Figure 4-5: Failure of complete preservative penetration in the heartwood samples. 51

Figure 4-6: The interaction between chemical, density and concentration for retention rate (kg/m^3) of CCA and DOT preservative treated samples. 52
Figure 4-7: Mean block shear strength and wood failure percentage values for CCA, DOT and control groups in accordance to EN 14080 (2013) average values requirements. 54
Figure 4-8: The effect of different CCA concentration levels in comparison to untreated on shear strength. 56
Figure 4-9: Effect of wood type in CCA treated and untreated samples on shear strength. 58
Figure 4-10: Penetration of adhesive in samples classified as heartwood 59
Figure 4-11: Effect of wood density on shear strength of CCA treated and control (untreated) samples. 60
Figure 4-12: A 3-way significant interaction between density, wood type and concentration for WFP in CCA treated and untreated blocks. 61
Figure 4-13: A 3-way significant interaction between concentration, wood type and density for shear strength (N/mm^2) in DOT treated and untreated blocks. 63
Figure 4-14: A 2-way interaction between wood type and concentration levels in DOT treated and untreated blocks for WFP. 65
Figure 4-15: Individual block shear strength and wood failure percentage values for CCA and DOT groups in accordance with EN 14080 (2013) individual values' requirements. 66
Figure 4-16: Mean shear strength for the different preservatives and treatment levels. 67
Figure 4-17: Mean WFP for different preservative and treatment levels. 68
Figure 4-18: CCA and control graph for total delamination. 71
Figure 4-19: Total delamination (%) of sapwood and heartwood. 72
Figure 4-20: Total delamination (%) of CCA and DOT. 74
Figure 4-21: Leaching out of DOT in delamination test blocks. 75

List of Tables

Table 2-1: Results of block-shear specimen tests (Gaspar, Cruz and Gomes, 2008). 21
Table 2-2: Delamination of southern yellow pine bonded with PRF. 27
Table 3-1: Experimental factors. 29
Table 3-2: Experimental design of CCA preservative groups. 30
Table 3-3: Experimental design of DOT preservative groups. 30
Table 3-4: Experimental design for control (untreated) groups. 30
Table 3-5: Density groups. 31
Table 3-6: Preservative solutions and targeted retention rates for CCA and DOT. 32
Table 3-7: Total number of samples per treatment. 37
Table 4-1: CCA groups mean shear, WFP and total delamination results. 43
Table 4-2: DOT groups mean shear, WFP and total delamination results. 43
Table 4-3: Control (untreated) groups mean shear, WFP and total delamination results. 44
Table 4-4: Retention rate of CCA and DOT preservatives for shear and delamination samples 47
Table 4-5: ANOVA table for retention rate results 49
Table 4-6: Minimum required values for wood failure percentage related to shear strength according to EN 14080 (2013). 54
Table 4-7: ANOVA shear strength results for CCA-treated and control (untreated) test blocks. 55
Table 4-8: ANOVA wood failure percentage results for CCA treated and control blocks. 61
Table 4-9: ANOVA results for shear strength of DOT treated samples. 62
Table 4-10: ANOVA table for WFP of DOT treated and control blocks. 64
Table 4-11: Benchmark values for delamination tests according EN 14080 (2013). 70
Table 4-12: ANOVA table for delamination of treated and untreated blocks. 70
Table 4-13: ANOVA table for DOT and control delamination test blocks. 73

List of abbreviations

CLT	Cross-Laminated Timber
MCA	Micronized Copper Azole
ACA	Ammoniacal Copper Arsenate
ACQ	Ammoniacal Copper Quat
CCA	Copper chrome arsenic
CCB	Copper chrome boric acid
1C-PUR	One component polyurethane
AWPA	American Wood Protection Association
LOSP	Light organic solvent preservative
EN	European standard
EPI	Emulsion Polymer Isocyanate
MUF	Melamine-Urea-Formaldehyde
PRF	Phenol-Resorcinol-Formaldehyde
WFP	Wood failure percentage
UF	Urea-Formaldehyde
RF	Resorcinol-Formaldehyde
PF	Phenol-Formaldehyde
SANS	South African National Standards
ASTM	American Society for Testing and Materials
ANSI	American National Standards Institute
LVL	Laminated Veneer Lumber
Glulam	Glued laminated timber
DOT	Disodium octaborate tetrahydrate
B.A.E	Boric Acid Equivalent
CSPG	Compression strength parallel to grain
MOR	Modulus of rupture
MOE	Modulus of elasticity
EWP	Engineered wood products
DSC	Differential scanning calorimetry

Chapter 1 : Introduction

1.1. Background to research question

The pressure for using sustainable building materials in the construction industry has led to increased use of alternatives that are environmentally friendly. Engineered wood products, such as glued laminated timber (glulam or GLT), laminated veneer lumber (LVL) and cross-laminated lumber (CLT) have become popular options, as they have lower environmental impacts (lower carbon footprint) when compared to mineral based building products such as concrete and steel.

In recent years, some engineered wood products such as LVL and CLT have grown from a novel invention to a much used product with building technology revolutionizing the use of massive timber in the construction industry (Muszynski *et al.*, 2017). Estimates from Europe indicated that 0.3 million cubic metres of mass timbers products had been used in buildings until 2010, with an estimated 1 million cubic metres which was forecasted for 2015 alone (Crespell and Gagnon, 2010; Kremer and Symmons, 2015). Global CLT production for 2020 was estimated to be close to two million cubic meters (Muszynski *et al.*, 2020).

In spite of the massive growth and use of engineered wood products that has been observed over the years, the adoption of these materials as building elements could prove to be a problem in some parts of the world when used for both interior and exterior applications, due to the different climatic conditions and degradation agents present. This is, because wood as a natural material is susceptible to biotic agents and natural elements, particularly in humid climates or environments where a moisture content of 20% or greater exist and temperatures ranging from 10˚C to 32˚C can occur. Such environmental conditions, if met, can accelerate the biological degradation of wood, as they are conducive for microbial growth and the harbouring of insects, fungi, and termites.

To date, most of the mass timber buildings have been constructed in locations with low decay and few insect hazards (Wang *et al.*, 2018) and to counter any possible biological risks (fungal or insect attack), biodeterioration has been typically controlled through recognized design principles and construction techniques, such as use of overhangs, flashings, ventilation and proper joint connection details (APA, 2013). But such design principles and construction techniques could prove to be ineffective in harsh weather conditions and where more severe biological decay agents exist.

Therefore, in high-risk areas, wood preservation remains one of the valuable alternatives to improve wood resistance and extend the service life through means of impregnation of chemicals into the wood cells - at levels which the chemical or preservative becomes toxic to decay agents. Through wood preservation, the fungus, insects, borers, and other decay agents can be restricted from accessing wood components (e.g. cellulose, hemicellulose, and lignin), making it unsuitable as a food source.

Treatment before gluing is the most effective way to do this, but some technical problems related to the gluing process must be solved (Gaspar *et al.*, 2010). Also, some engineered wood products, such as CLT and very large GLT beams cannot be treated after production due to dimensional constraints of treatment facilities. However, the presence of wood preservative deposits in wood have been reported to adversely affect the bond performance of laminates. According to Lim et al. (2020), wood preservative deposits can physically and chemically block surfaces where the intermolecular forces of adhesive bonding develop, they also reduce wettability and surface energy of the wood, increase contact angle, which in turn reduces adhesion, penetration

and spreading of the adhesive and may also alter the curing rate of adhesive. This is evident in a study conducted by Özçifçi (2006) where it was reported that the metallic deposits or active ingredients (Cu, Cr, As) contained in copper chromium arsenic (CCA) preservative, significantly affected the shear strength of glue bonds in solid wood samples. Similarly, Vick et al. (1990) reported that non-acidic borate-based waterborne preservatives, including ammoniacal copper borate (ACB), ammoniacal pentaborate (AP) and disodium octaborate tetrahydrate (DOT) caused poor bonding even at the lowest retention level. Vick (1999) and Tascioglu et al. (2003) theorised that these deposits reduce contact on a molecular level between the adhesive and lignocellulosic wood material and lead to weaker bonds. On the contrary, Ozdemir et al. (2015) found that boric acid and copper azole provided increased adhesion strength.

Since literature presented contradicting findings on the effect of wood preservatives on bond line performance and with very limited research done on the compatibility of PUR adhesive to CCA and DOT-treated *Pinus patula,* it was decided that additional research is required. The experimental work of this research aimed at evaluating the effects of two waterborne preservatives (copper chromium arsenic and borate-based preservative disodium octaborate tetrahydrate) at different concentration levels (CCA: 2% and 4%; DOT: 1.67% and 3.33%) on the performance of 1C-PUR adhesive bond lines in *Pinus patula*. The effects of wood properties – density and heartwood to sapwood ratio were also investigated as they have been reported by as factors that may also influence the bond performance (Vick, 1999; Kaygin and Tankut, 2008; Hunt *et al.*, 2019). The performance of PUR adhesive bonds was evaluated and measured through standardised test methods including shear strength, wood failure percentage (ASTM D905, 2008; EN 14080, 2013) and resistance to delamination by accelerated exposure according to SANS 10183-4-2 (2009).

1.2. Problem statement

Wood by its nature is susceptible to deterioration when exposed to fluctuating climatic conditions that harbour or favour the survival and growth of wood decay agents, such as fungus and insects. Some of the countries where mass timber structures have recently been implemented have tropical climate, with high temperatures and humidity as well as severe biodegradation hazards (Oliveira et al., 2018). As such, wood structural components located in those regions are more susceptible to building pathologies caused by bio-deterioration, than they are in dry or cold climates (Oliveira et al., 2018).

Therefore, to improve wood resistance against biodegradation and increase the product service life, the adoption of preservation treatment must be explored for engineered wood products. For some products like CLT the final product dimensions make post-manufacturing treatment impossible and, therefore, laminates need to be treated before the adhesion process. In some cases, pre-treatment of laminates can also simplify the production process of products like glulam.

However, some wood preservatives may present complications as they can interfere with bond formation and lead to poor bond performance (bond durability and strength). Hence, this research aims to evaluate and determine whether wood preservatives (copper chromium arsenic and disodium octaborate tetrahydrate) affect the bond strength and durability of *Pinus patula* structural laminates, bonded with a one 1C-PUR adhesive. This research will also investigate the influence of wood properties (e.g. density, sapwood/heartwood) on preservative retention.

1.3. Objectives

The objectives of this study were as follow:

- Evaluate the effect of chromium copper arsenic (CCA) and disodium octaborate tetrahydrate (DOT) wood preservatives on adhesive bond performance (shear strength, wood failure and delamination) of *Pinus patula* laminated with a one component PUR adhesive;
- Evaluate the influence of *Pinus patula* wood properties (sapwood to heartwood ratio, and density) on retention and bond quality.

1.4. Brief Chapter Overview

This thesis consists of five chapters and report on research related to the influence of CCA and DOT wood preservative treatments on 1C-PUR adhesive bonded *Pinus patula* structural laminates. Chapter 1 is the introduction, which outlines the background, problem statement and research objectives. Chapter 2 provides what is currently known about the research topic (literature review). Chapter 3 illustrates the material and methods used to conduct the research and realise the objectives of the thesis, and in chapter 4, results are presented and discussed. Chapter 5 is the conclusion and recommendations for future research.

1.5. Approach and procedure

In order to evaluate the bond strength and quality of CCA and DOT treated wood laminates, bonded with a 1C-PUR adhesive - samples of *Pinus patula* lamellas were impregnated with CCA and DOT preservatives at different concentration levels, using a modified empty cell process. The wood samples were bonded with a 1C-PUR adhesive afterwards to produce laminates. Bond performance tests - shear strength (ASTM D905, 2008; EN 14080, 2013), wood failure percentage, and delamination (SANS 10183-4-2, 2009) were used to determine whether wood preservatives alter the structural performance of treated laminates.

It should be noted that in the research specimens for delamination and shear testing was glued in the parallel-to-grain direction (as with glulam). The reason was that delamination testing standards for CLT are still subject to research and has been criticized for being too harsh whereas glulam delamination testing standards are well established (Betti *et al.*, 2016; Knorz, Torno and van de Kuilen, 2017; Dugmore, 2018).

1.6. Limitations and constraints of the experiment or study

A surface characterization by means of various analytical techniques (ESEM and XPS), which was outside the scope of this research, would have perhaps provided a better understanding on the surface properties of treated wood, such as roughness, contact angle, wettability, surface energy and pH changes before bonding.

Secondly, the inclusion of other structural adhesives, such as phenol resorcinol formaldehyde (PRF) and melamine urea formaldehyde (MUF) would have enabled the comparison of the different adhesives, on how they perform in the presence of CCA and DOT.

Also, extractive characterisation (composition, type, quantity and pH of extractives, content of fatty acids etc.) in heartwood samples would have uncovered and assisted in identifying, which extractives might have improved the bond strength in heartwood specimens of *Pinus patula*.

Chapter 2 : Literature Review

2.1. Engineered wood products

While wood as the most important renewable structural material has always been an essential part of the built environment, the importance of using sustainable construction materials is increasing the rising global demand for housing and an increase in the understanding of the impacts the built environment has on climate change (Connolly *et al.*, 2018). This has led into an increase in utilisation of engineered wood products, such as Glulam, CLT and LVL as the world looks to reduce the use of traditional building materials (e.g. steel, concrete etc.) and adopting or opting for more environmentally friendly and sustainable materials that contribute to CO_2 emission reduction and storage.

Laminated timber products are often described as a group of engineered wood products manufactured from multiple layers of wood boards with an adhesive under pressure. These products come either as a honeycomb system, using primarily cross-laminated timber (CLT), or as post and beam construction using a mix of CLT, glue laminated timber (Glulam), [NLT(nail laminated timber)], finger jointed solid timber and laminated veneer lumber (LVL) (Crawford and Cadorel, 2017). Their respective structures and application are shown in Figure 2-1. The ease to assemble, reduced noise, natural beauty, opportunities for prefabrication on site, avoidance of fossil-fuel intensive materials, excellent seismic performance are some of the attractions towards the use of engineered wood products (Wang *et al.*, 2018; Gong, 2019).

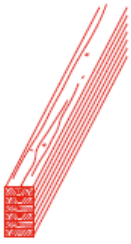
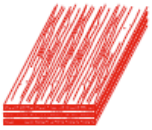
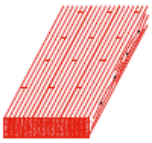
Engineered Timber Product	Parallel Strand Lumber (PSL)	Laminated Veneer Lumber (LVL)	I-Joist	Glulam	Structural Insulating Panel (SIP)	Cross Laminated Timber (CLT)	Brettstappel
Typical Detail							
Application	• Beams • Columns	• Beam • Columns • Cord	• Joist • Beam	• Beam (Long span) • High Loading	• Roof • Wall • Floor	• Roof • Wall • Floor	• Roof • Wall • Floor
Usage	Interior	Interior	Interior	Interior / Exterior	Interior	Interior/ Exterior	Interior/ Exterior

Figure 2-1: Common engineered wood products (Ramage et al., 2017).

Most engineered wood products are produced from softwood timber, with hardwood species rarely being used for structural purposes. This is mainly due to the tendency of hardwoods to check and split as well as the low dimensional stability of the wood, which causes the boards to warp extensively and hence cannot comply with the building standards requirements (Crawford, (2010) cited in Pröller, (2017)). Typical softwoods such as spruce (*Picea spp*.), lodgepole pine (*Pinus contorta*) and Douglas fir (*Pseudotsuga menziesii*) are commonly used for the construction of mass timber structures in Europe and North America (Dugmore, 2018).

Concerns

Although, the development of engineered wood products has the potential to revolutionize the use of timber in buildings (Wang *et al.*, 2018), the susceptibility of these products to biological agents (e.g. fungus, insects, rots, moulds and ultraviolet light) is still a global concern. Constructing mass timber structures without any wood preservative presents the opportunity for building pathologies to degrade the structure over time. These building pathologies can interfere with the structural integrity of wood, as cellulose, hemicellulose and lignin components in wood remain accessible. The biodeterioration of mass timber structures becomes more prevalent in tropical climates, since in such locations, the environmental conditions are more aggressive relating to the biodiversity of the pathogens, temperature and humidity, than in cold and dry climates (Oliveira et al, 2018). Such environmental conditions overtime can interfere with the structural integrity of the mass timber structure and lead to structural failure. Failure to deal with these conditions may halt their development and prevent wood from replacing materials that are based on unrenewable resources (Shams, Yano and Endou, 2004).

However, making use of wood preservatives, the service life of EWP can be drastically improved as they provide resistance against insects, termites and fungi and moreover provide wood with the ability to inhibit photo-induced degradation. With the expected growth in use of engineered wood products, wood preservation industry needs to be considered and remain a viable option to protect wooden structures from decay agents.

2.2. Wood preservation

Wood preservation offers the opportunity to improve wood resistance to wood deterioration agents through the impregnation of wood preservative chemicals (into the wood cell). This extends the service life of wood products and enhance the ability to inhibit photo-induced degradation. According to Environment Canada (2013), wood preservation enhances the lifetime utility of wood by a factor of 5 to 10 or more, depending on the species, end use and efficacy of the treatment.

2.2.1. Wood preservatives

Wood preservatives are mainly in liquid form and rely on solvents to carry the toxic chemical into the wood cells during impregnation. They are mainly divided into three primary groups, namely, water-borne preservatives (e.g. *CCA, Borates, Copper azole, ACQ etc.*), oil preservatives (e.g. *Creosote*), and light organic solvent borne preservatives (e.g. *TBTN-P, Azole-permethrin*). The effectiveness of these preservatives varies greatly and depends not only upon its composition, but also upon the quantity (retention rate) injected into the wood, wood cell structure, chemical inclusions within cells, density, impregnation technique and post-treatment procedures (*Wood Preserving*, no date).

Below is a detailed description of the waterborne preservatives used in the experimental work of this research:

2.2.1.1. Copper-Chromium-Arsenic – Fixed preservative

CCA preservatives are widely used for the treatment of various types of wood products, which are made from a wide range of wood species. The combination of being highly effective against the broad spectrum of biological agents and being highly permanent (i.e. fixed) make them a unique option for wood preservation (Aston, 1985). The CCA preservative consists of three inorganic compounds, in the form of oxides or salts,

that each act as a nemesis to decay agents. The copper (Cu) is a primary fungicide, whilst arsenic (As) is an insecticide. The chrome (Cr) acts as a fixing agent, reacting in the presence of wood cellulose to render the copper and arsenic chemicals insoluble (SAWPA, n.d.).

CCA preservatives can be classified into three formulation types as specified by AWPA (1991):

- *A* (CuO 18.1%, Cr_2O_3 65.5%, As_2O_5 16.4%),
- *B* (CuO 19.6%, Cr_2O_3 35.3%, As_2O_5 45.1%),
- *C* (CuO 18.5%, Cr_2O_3 47.5%, As_2O_5 34%).

These three formulations differ in the relative proportions (oxide basis) of chromium, copper, and arsenic (Lebow, 1996).The copper (CuO) content of the three CCA formulations is similar while large differences lie in the balancing between chromium and arsenic. The use of CCA-B type is often confined to field and remedial treatments, while CCA-A has high chromium content with relatively few treaters using it. CCA-C type is the most used for wood preservation, as the formulation appears to offer the best combination of performance and leach resistance (Lebow, 1996).

However, CCA is currently facing severe restrictions in the US, Europe and in other parts of the world but is still widely used and considered one of the most effective acidic waterborne preservatives in the world (Tascioglu, 2002).

2.2.1.2. Borate compounds – Non-fixed preservatives

Borate compounds as wood preservatives are known to have several advantages including, providing resistance against insects and fungal degradation, low mammalian toxicity, non-corrosive on metal joints/tighteners and absence of colour and odour after treatment (Özçifçi, 2006). Some boron compounds also have the ability to act as fire retardants, when a phosphate-based fire retardant is added. These inorganic salts release acid when the temperature is elevated, which decreases the flammable volatiles and increase the char rate in wood. Colakoglu et al., (2003) also found that when wood is treated with inorganic salts, such as boric acid, diammonium phosphate, and ammonium sulfate, these chemicals alter the combustion properties of wood, increasing the amount of char and reducing the amount of volatile.

Boron compounds are often diffusible and can be applied in species that are difficult and achieve excellent penetration. Even when not applied on the whole cross section, they can redistribute by diffusion if sufficient moisture is available in wood (Freeman et al. 2009).

However, owing to the water solubility of borates, these preservatives tend to be mostly suitable for interior use timber (or areas of less moisture exposure), or unless combined with an appropriate water-repellent system that can provide long-term protection against leaching when used in exterior above-ground H3 conditions (SANS 10005, 2016).

Over the years several borate-based wood preservatives have been developed including sodium tetrahydrate, sodium pentaborate, zinc borate, borax ($Na_2B_8O_{13}\cdot H_2O$), boric acid (H_3BO_3), disodium octaborate tetrahydrate ($Na_2B_8O_{13}.4H_2O$), a mixture of borax and boric acid and a polyborate deviation that contains emulsified wax. For comparison purposes among borates, standard units known as Boric Acid Equivalent (B.A.E) or Boric Oxide (B_2O_3) are often used to compare the efficacy of borate-based compounds. The B.A.E or boric oxide compares the amount of boric acid that could be formed from the primary or subject compound,

as all borates convert to boric acid when they dissolve in acidic media such as in wood (pH 4 – 5) (Freeman et al. 2009).

Disodium octaborate tetrahydrate (DOT) has gained much commercial popularity, mainly due to the high-water solubility of DOT, which allows the use of higher mass concentrations and increasing mobility in wood. On the contrary, the high solubility of DOT is often a disadvantageous attribute with regards to leaching, as it tends to lose more boron when compared to other boron compounds. DOT often contains more boron per unit mass (20.9%) followed by boric acid (17.48% boron), and borax (11.4%) (Freeman et al., 2009). In spite of the high percentage of boron contained in DOT, the effectiveness of boron compounds mainly depends on the quantity or amount (mass concentration) of boron compound applied in wood, whether boric acid, borax or DOT.

In an attempt to reduce the leaching of boron in boron compounds and expand its use to exterior applications, more complex formulations have been developed in combination with copper, chromium, and quaternary ammonium. Such combinations have produced wood preservatives such as CCB (copper chromium boron). Selamat et al., (1993) evaluated the effectiveness of CCB as a wood preservative when compared to CCA – results showed that CCA and CCB preservatives gave almost the same degree of protection at 6% of solution strength. However, there was more severe loss of boron from CCB treated timber when compared to arsenic from CCA treated timber. This is mainly due to the boron in CCB preservative that is largely unfixed in the wood and leaches out when the timber is exposed to rain and ground contact (Selamat et al., 1993), whilst arsenic in CCA remains fixed, which makes CCA a highly leach resistant preservative.

2.2.2. Impregnation techniques

There are several impregnation techniques that can be employed to ensure the transportation of preservative active ingredients into the wood cells. These impregnation techniques can be generally classed into two groups: pressure processes (full cell and empty cell) and non-pressure processes (brushing, spraying, dipping, soaking, diffusion). The selection of the impregnation technique often depends on the key indicators of impregnation: targeted retention rate (kg/m^3) and penetration (mm) and other factors, which include hazard class-exposure, wood species, and size of product, permeability, and moisture content.

Pressure impregnation processes are generally the best and most common techniques used as they achieve a much deeper and uniform penetration in a relatively short period of time as compared to non-pressure processes. Pressure impregnation processes (Figure 2-2) generally operate on the same principle and differ on the details of application. They occur in an enclosed treating cylinder where wood is impregnated with a preservative solution at high pressure.

For instance, the empty cell process is designed to obtain deep penetration with a relatively low net retention of preservative (Groenier and Lebow, 2006). The final weight of empty cell treated wood is reduced when compared to the full cell. The empty cell process has two treating processes, namely: Rueping process and Lowry process, which operate on similar methods as the full cell (Bethel) process except for the initial vacuum.

Rueping: wood (charge) is placed in an enclosed cylinder and an air pressure (generated by a compressor), higher than atmospheric pressure is applied. The treatment then continues as with the full cell

process, but the amount of preservative removed (as the air compressed in the cells expands) is greater than in the Lowry process (Milton, 1995).

Lowry process wood (charge) is placed in an enclosed cylinder and the preservative is pumped into the cylinder, with no air allowed to escape. After the cylinder is filled with the preservative, pressure is applied and maintained at maximum pressure (the air in the cylinder and wood cells is compressed and its occupation decreases into smaller space). The process then continues exactly as the full cell process, but the air compressed inside the wood expands when the pressure is released, thereby forcing some preservative out of the cells and eliminating overloading (Milton, 1995). The end result is that many cells are “lined” with preservative rather than “filled” (Milton, 1995).

The Lowry process has the advantage that equipment for the full-cell process can be used without other accessories that the Rueping process usually requires, such as an air com- pressor, an extra cylinder or Rueping tank for the preservative, or a suitable pump to force the preservative into the cylinder against the air pressure (Forest Products Laboratory, 2010).

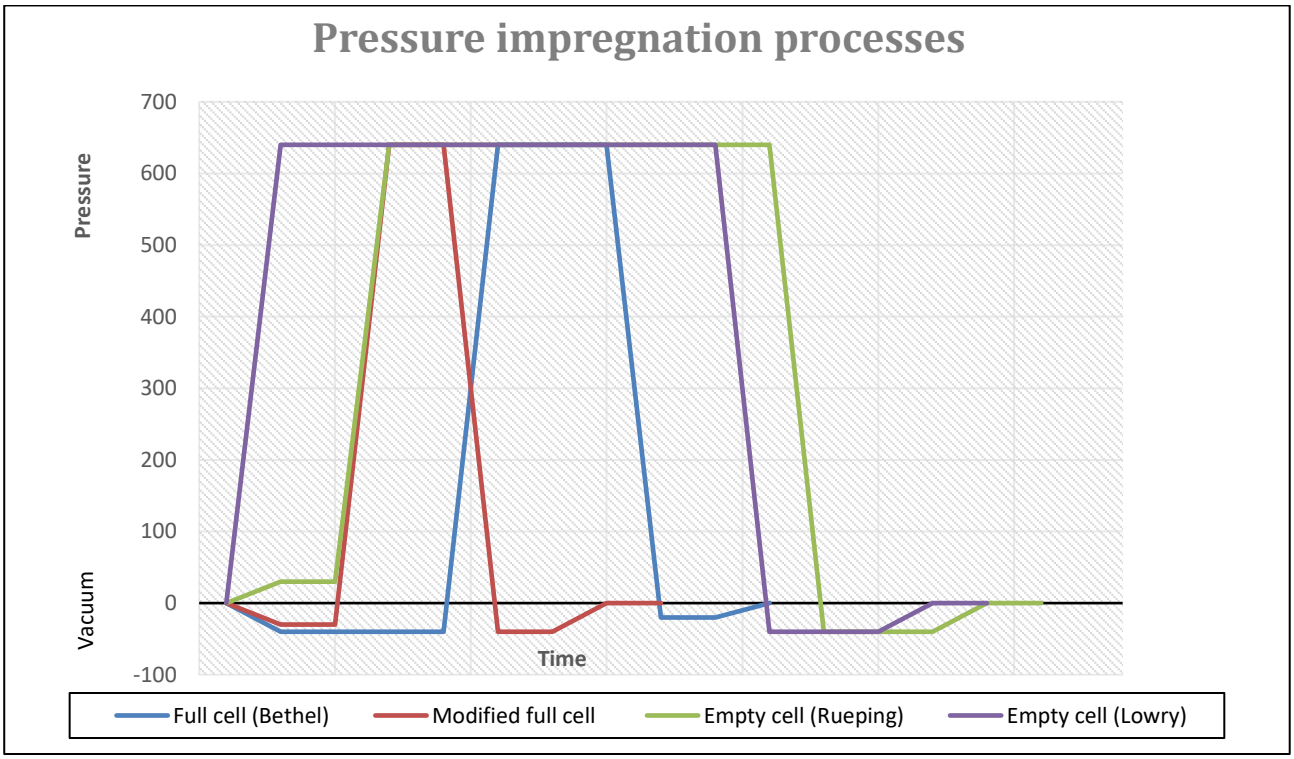


Figure 2-2: Pressure treating cycles.

2.2.3. Absorption of preservatives in softwood

One of the most important aspects of wood as far as impregnation is concerned is related to its porosity and how internal cavities or lumens at the microscopic level communicate with each other (Olsson *et al.*, 2001). The porosity of wood is determined by a combination of several factors including latewood/earlywood proportion, density, sapwood/heartwood ratio, type of cells, cell size, bordered pits membrane and aspiration, number of pits and chemical inclusions (extractives). In softwoods preservatives flow occurs by means of the vertical fibers tracheids and the horizontal ray tracheids (Milton, 1995), and these softwood cells virtually make

up the total wood volume. A significant obstacle to liquid flow in softwoods is often related to the aspiration process, which affects the capillary flow, and the structure of the bordered pit membranes (Olsson *et al.*, 2001). As Figure 2-3 shows, the liquid would mainly enter on the *tangential face* (ray tracheids) and *transverse face* (tracheids) of the softwood cube, while the radial face – which has unexposed cells, will not be able to absorb the preservative. The hollow tube tracheids on the *transverse face* will absorb the preservative since their end grain is exposed. Their small diameter actually encourages "sucking-in" of the liquid by capillary action […] (Milton, 1995) and as the preservative enters the tracheids, it will pass through the pits in the cell wall into adjoining tracheids. In the *tangential face*, the ray tracheid cells are also capable of capillary absorption of preservatives.

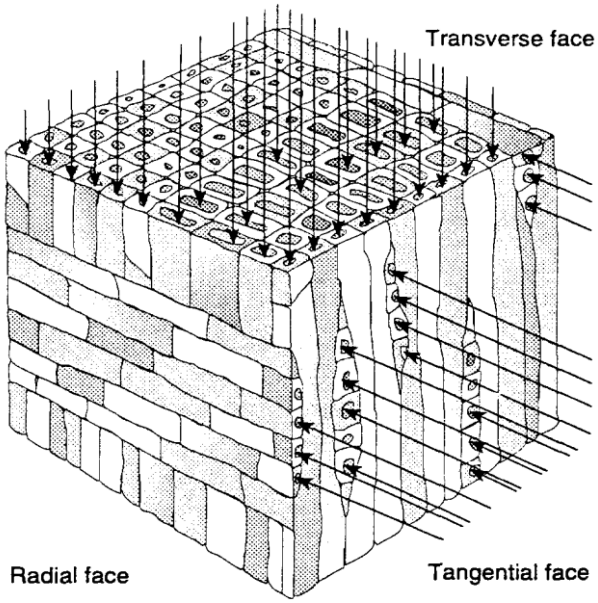


Figure 2-3: Softwood porosity structure (Milton, 1995).

2.2.4. Effect of preservatives on chemical properties of wood

When wood has been impregnated with preservatives, such as CCA, the cellular surfaces of wood are thoroughly covered with microscopic-size deposits of mixtures of chromium, copper and arsenic oxides that are physiochemically fixed to cell walls (Vick, 1999). The presence of these insoluble metallic deposits is so pervasive that intermolecular forces of attraction that normally act between polar wood and adhesive are physically blocked (Vick, 1999).

In a review conducted by Winandy (1987), it identified that some waterborne preservatives (e.g. CCA) were shown to generally reduce the strength properties of wood, as many of the metallic oxides used in waterborne preservatives formulations (mainly containing high chromium percentage) do react with the cell wall components by undergoing hydrolytic reductions upon contact with wood sugars. In this process, known as fixation/precipitation period, the metals are reduced to less water-soluble forms by oxidizing the wood cell-wall components (Yildiz *et al.*, 2004). During the fixation period, CCA metallic ions tend to react with cellulose and lignin forming complexes. Copper (Cu^{2+}) metallic ions form complexes with cellulose and lignin and are

physically absorbed on wood (Vick and Christiansen, 1993), while chromium arsenate($CrAsO_4$) complexes with lignin and precipitates on cellulose and $Cr_2(OH)_4CrO_4$ also precipitating on cellulose (Vick and Christiansen, 1993). The metallic salts of CCA are known to be reactive and may promote corrosion of mechanical fasteners.

On the other hand, Winandy and Rowell (2009) found that some waterborne preservatives (e.g., ACQ, CA, borates) become insoluble as treated wood dries, which dehydrates the preservative complex within the wood. This reaction is known as immobilization, as the cell wall is not directly affected and subsequently strength is virtually unaffected. However, the insolubility of such preservatives or failure to react with the cell wall, often leads to leaching of the preservative whenever exposed to frequent wetting.

2.2.5. Effect of preservatives on mechanical properties of wood

Modulus of elasticity (MOE) and modulus of rapture (MOR) are mechanical properties, which are the primary criteria for the selection and design of engineered wood products. Therefore, whenever wood building components are treated with wood preservatives, they should more or less possess the same mechanical properties as the untreated building components.

According to Yildiz *et al.* (2004) the effects of waterborne wood preservatives on mechanical properties have been shown to be directly related to several key wood material factors and pre-treatment, impregnation technique, preservative chemistry, solution concentration level and post-treatment factors.

To determine the effects of wood preservatives on mechanical properties, Yildiz *et al.,* (2004) investigated the effects of CCA and other new wood preservatives on modulus of elasticity (MOE) and modulus of rupture (MOR) of yellow pine (*P. Sylvestris*) sapwood. The results showed a decrease in MOE and MOR on wood samples treated with CCA, ACQ-2200 and Tanalith E 3491 (except MOR for Tanalith E 3491). In addition, as the concentration levels increased, the MOR and MOE decreased. However, with ACQ 1900 and Wolmanit CX-8, results showed an increase in MOR and MOE as the concentration levels increased.

Simsek et al., (2010) conducted a similar study where mechanical properties (MOR) of Oriental beech and Scots pine treated with 3 environmentally friendly borate-based preservatives (SFB, AFB, APB) were determined. The results showed that borate treatments caused a decrease on MOR . Furthermore, the results also revealed that the higher the concentration levels of borates, the lower MOR of wood becomes. Simsek et al., (2010) concluded that, the decrease in mechanical properties of wood treated with borates, might be due to the fact that borates increase the rate of hydrolysis in the wood, thereby causing loss in strength.

Winandy and Rowell (2009) reported that wood preservative chemicals can swell, hydrolyze, pyrolyze, oxidize, and, in general, depolymerize wood polymers, causing a loss in strength properties. They further highlighted that in some cases, the loss in mechanical properties caused by wood treatment may be large enough that the treated material can no longer be considered the same as the untreated material. FAO (1986) further highlighted that the high pressures applied during the pressure impregnation process can be a major factor which may affect the strength of timber as they can cause the wood cells to collapse especially in low density timber.

In addition, most water-borne preservative salts increase the hygroscopicity of the wood, which causes an increased EMC, which further influences strength (Winandy and Rowell, 2009).

In contrast, Nicholas and Preston (1988) reported that borate-based preservatives do not degrade wood and in fact a slight increase in MOR values have been observed.

2.2.6. Effect of preservatives on surface properties of wood

Because adhesives bond by surface attachment, the physical and chemical conditions of the wood's surface are extremely important to satisfactory bond performance (Frihart and Hunt, 2010). Therefore, in order to achieve higher adhesion strength and maximal surface interactions, the surface energy of wood and adhesive should be almost equal. However, wetting the surface of treated wood can be a challenge due to the modification of the wood's surface and chemical contamination. For example, the pH of wood becomes acidic instantly as soon as it comes into direct contact with the acidic CCA preservative. This is due to the ion exchange and adsorption reactions that occur between the metals and wood (Vick and Kuster, 1992), which effects the adhesion.

According to Frihart (2003) adhesion is diminished when the wood surface is covered by chemicals, whether natural oils and adhesives or added chemicals (wood preservatives or fire retardants). Frihart and Hunt (2010) highlighted that wood preservation leads to the deterioration of the wood surface and as a result, this causes unevenness on the wood surface, causes air pockets and blockages which can prevent complete wetting by the adhesive and introduce stress concentrations when the adhesive has cured.

Furthermore, Frihart (2004) also highlighted that many wood treatments tend to reduce the water adsorption of the wood, which is a good property for decay resistance, however, this causes the wood to have poor surface wettability properties and reduced surface energy. Most wood adhesives are water based; thus, they need high wood surface energies to be able to wet and penetrate the wood (Frihart, 2004). As such, in treated wood, the adhesive is often slow to wet the wood, which causes the adhesive to cure before it flows into the cell cavities/lumens and ultimately leading to weak or poor bonds.

Maldas and Kamdem, (1998), performed a surface characterization on CCA treated red maple. The contact angle, which measures the wettability of solid surfaces by liquid, was one of the indicators used in the experiment. For CCA treated wood, (Figure 2-4) a higher contact angle was observed with distilled water (wetting liquid), which suggests that CCA treated wood surfaces have poor wettability properties as also highlighted by Frihart and Hunt (2010) and Tascioglu et al., (2003).

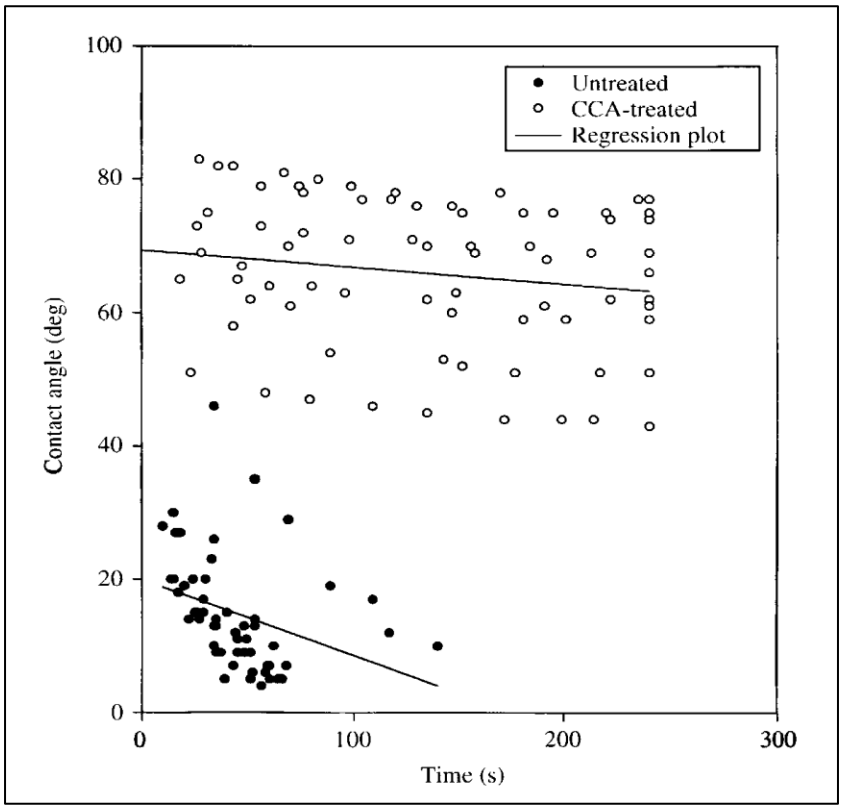


Figure 2-4: Time-dependent contact angle with all the replicate data points for untreated and CCA-treated wood with distilled water used as a wetting liquid (Maldas and Kamdem, 1998).

They concluded that the higher contact angles on CCA treated wood surface was caused by the presence of the wood preservative deposits. The deposition of As, Cu, and Cr oxides causes the wood surface to become rougher, less polar, hydrophobic, and acidic. The contact angle also influences the rate that the adhesive advances through a capillary such as a lumen (Hunt *et al.*, 2019).

2.3. Wood adhesives

Wood adhesives used for the assembly of EWP should have the ability to withstand extreme weather conditions and withstand high loads as the safety of inhabitants is at stake. For this reason, adhesives must satisfy the requirements of structural adhesives standards, such as ANSI 405 (2013), SANS 10183-2 (2014), and EN 302 (2013).

2.3.1. One component polyurethane adhesive

With various adhesives introduced into the production of engineered wood products, this section discusses the properties of PUR adhesive system used for modern timber structures.

Adhesives for load-bearing timber structures, such as Glulam and CLT, must generally resist high static and dynamic mechanical loads, as well as high stresses due to the swelling or shrinking of wood resulting in increased elastic and even plastic deformations (Clauß *et al.*, 2011). Over the years several adhesives (UF, MF, MUF, PR, RF and PRF) have been developed and improved for structural timber, with 1C-PUR being recently developed and accepted for use in timber structures. The one component polyurethane adhesive has been promoted as a waterproof adhesive and is suitable for exterior and interior applications (Vick and Okkonen, 2000). The use of 1C-PUR in the production of engineered wood products has continuously

increased as it offers several advantages, such as reduction in press time, contains no formaldehyde, 100% solid content, no curing agent required, creates a clear bond line and has a fast-curing rate at room temperature.

Because of their hardening chemistry, one-component PUR adhesives are also suitable for gluing timber at high moisture content (MC), which is known as wet or green gluing (Serrano and Kallander, 2005), while traditional urea-formaldehyde based aminoplasts adhesives (UF, MF, MUF) have shown a tendency to hydrolyze under the influence of increased moisture (Lehringer and Gabriel, 2014). In addition, 1C PUR bond lines have shown an increased ductility, a characteristic that differs significantly from aqueous or formaldehyde-based adhesives, which are usually more brittle with a higher modulus due to a high crosslink density (Pröller, 2017).

However, 1C-PUR tends to produce a slight foam during hardening as it is reactive towards moisture and creates a weak point along the glueline (Yusof *et al.*, 2019; Vick and Okkonen, 1998). Yusof *et al.*, (2019) investigated the bond integrity of CLT fabricated from *Acacia mangium* wood by using PRF and PUR as adhesives. The CLT panels bonded with PRF, showed superior properties in terms of shear strength and wood failure percentage when compared to CLT panels bonded with PUR. The superior properties of PRF were attributed to better gap-filling properties.

Furthermore, 1C PUR adhesives are characterized by a significantly lower stiffness and hardness compared to amino- and phenoplastic resins, but absorb much more deformation energy and show ductile failure behaviour leading to lower wood failure (Clauß *et al.*, 2011; Pröller, 2017).

Lim, Tripathi and Tang (2020) tested the bonding performance of three adhesive systems (PUR, MF, RF) on CLT treated with micronized copper azole type C, at two retention levels (1 kg/m^3 and 2.4 kg/m^3). The delamination rates of the treated specimens assembled using MF and RF increased with the preservative retention level, while PUR achieved delamination rates less than 1% to the laminations treated at both levels. The lower delamination rate of PUR was attributed to its capability of absorbing additional energy upon deformation, a favourable characteristic when wood is exposed to frequent wetting and drying cycles.

Vick and Okkonen, (1998), compared four commercial one-component PUR adhesives with one PRF adhesive. They found that the dry strength of the PUR adhesives is at least as high as that of the PRF. After the water saturation process, the wet shear strengths were still statistically comparable. However, measurements of wood failure indicated that polyurethane bonds were not equivalent, and a moderately severe delamination test indicated varying levels of water resistance among the polyurethanes.

Sikora, McPolin and Harte (2016) compared the durability of PUR and PRF at different clamping pressures. The results showed higher shear strength values for PUR specimens, while PRF specimens demonstrated superior durability characteristics in the delamination tests.

Furthermore, Maldas and Kamdem (1998) and Lisperguer *et al.* (2005) reported that, many conventional wood adhesives, such as PF, UF and PRF, do not adhere to preservative-treated wood well enough to meet industrial standards for resistance to delamination. In addition, the rigidity of PRF adhesives limit its ability to respond to moisture induced dimensional changes such as swells in wood and potentially creates large stresses at the interface.

2.3.2. Penetration of adhesives

Unlike any other substrate, wood is an anisotropic material which is relatively easy to bond, as it contains complex multi-cellular anatomical features which provide a pathway for the flow of adhesives. In softwood species adhesives penetrate and flow through the tracheids voids/lumen and ray tracheids and further distribute through the interconnected pits to develop molecular interactions and provide mechanical interlocking (Frihart and Hunt, 2010).

The manner of penetration of adhesives in wood may be categorized into two different phenomena's: gross penetration and cell wall penetration. *Gross penetration* is described as the flow of the bulk of the adhesive, whether on surface as in wetting or flow into the wood to fill the cell lumens. This phenomenon is described by hydrodynamic flow and capillary action. The hydrodynamic flow is a result of the application of an external force (clamp pressure) on wood substrates to be bonded. This forces the adhesive to penetrate the wood surface and fill the cell lumens/voids, as it follows a path of least resistance (Kamke and Lee, 2007). The capillary action is the net result of wetting of internal surface and the surface tension of the liquid (Kamke and Lee, 2007). As such, this makes the character or properties of the internal surface (lumen wall) just as important as the external surface, as it also affects the penetration of the adhesive.

Cell wall penetration, occurs when the adhesive diffuses into the cell wall or flows into micro fissures (Kamke and Lee, 2007), provided that the adhesive has a low molecular weight. This infiltration of the cell wall is controlled by a molecule's hydrodynamic volume and solubility parameter (Frihart, 2009). Once the adhesive penetrates and fill the cell lumens and cell walls, the wood-adhesive bond forms as the liquid adhesive changes its state and solidifies. The applied adhesive changes from liquid to solid by one or more of three mechanisms: (a) loss of solvent from adhesive through evaporation and diffusion into the wood, (b) cooling of a molten adhesive, or (c) chemical polymerization into cross-linked structures that resist softening on heating (Frihart and Hunt, 2010).

2.4. Factors influencing bond formation and performance

There are several intertwined factors that may influence bond strength and quality. These factors include both wood and adhesive and processing related properties shown in Figure 2-5. All these factors (see Figure 2-5) act and interact when the adhesive cures, to determine the final mechanical properties, such as bond strength, stiffness, fracture energy, fracture behaviour and long-term properties (Sterley, 2012).

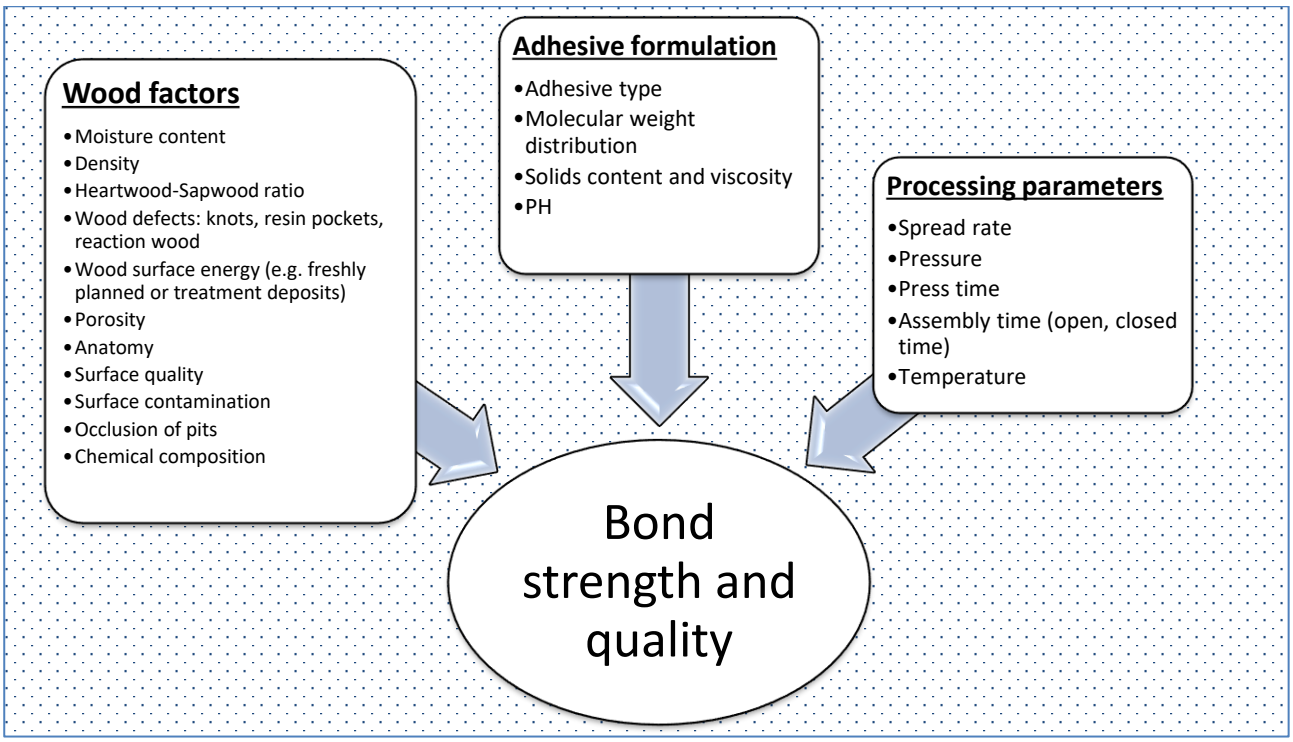


Figure 2-5: Factors affecting bond strength and quality.

Below is a summary of some of the common factors which should be considered when bonding engineered wood products, especially treated wood products:

2.4.1. Density

Density is one of the major factors of wood that can affect bond formation and mechanical interlocking. The density of wood represents a combination of anatomical characteristics, which can be described as the amount of material in the cell wall (thickness), cell wall thickness, cell lumen, heartwood/sapwood ratio, juvenile wood, and latewood/earlywood proportion (Kamke and Lee, 2007; Malan, 2011; Hunt *et al.*, 2019).

Often high density is a desirable property in wood as it is positively correlated to wood strength and stiffness. This is because the thick-walled cells are capable of withstanding much greater stress (Vick, 1999) and can carry more load (Frihart and Hunt, 2010) than thin-walled cells of low density. Vick (1999) reported that the strength of adhesive bonds to wood increases with wood density. In contrast, high density wood species can be extremely difficult to bond due the small cell lumen openings/thick cell walls, which can restrict adhesive penetration and severely compromise the depth of mechanical interlocking to two cell deep (Dugmore, 2018). This can lead to squeezing out of adhesive or leaving a large area of the glueline exposed to moisture when the glueline is too thick and severely compromising the mechanical interlocking between wood substrates. This phenomenon was reported by Pröller (2017) were 1C-PUR adhesive showed poor adhesion quality and high delamination values when applied in dry, high-density wood. In addition, high density wood also has increased shrinkage and swelling, and such stresses may initiate/cause bond failure in the gluelines when bonds are exposed to moisture changes (Hunt *et al.*, 2019). On the other hand, low density wood is usually easy to bond, but if the timber is too porous, too much adhesive can be absorbed by the pores, resulting in a starved bond line and reduced bond strength (SANS 1460, 2015).

2.4.2. Heartwood

Generally, the heartwood has a low permeability due to the small pore cell sizes, the irreversible nature of pit aspiration in the heartwood, the amount and type of extractives deposited on pit membranes during the formation of heartwood (Tripathi, 2012).

In terms of bonding, the presence of extractives and aspiration of pits in heartwood may inhibit the interaction between the wood and adhesive and thus cause bonds of weaker strength (Kaygin and Tankut, 2008; Clauß *et al.*, 2011; Roffael, 2016). These extractives can also alter the curing of adhesives and acidity of wood. The acidity of wood extractives can accelerate the hardening speed of acid curing urea-formaldehyde adhesives (UF-resins) and decelerate that of alkaline phenolic adhesives (PF-resins) (Roffael, 2016). According to Clauß *et al.*, (2011), the high level of extractives in heartwood may also reduce the wettability of the surface as well as the flow and penetration of the adhesive. Nonetheless, the high levels of extractives (such as tannin and terpenoids) in heartwood provide natural resistance against degradation from decay organisms in most species.

Hse and Kuo (1988) reported that during the drying process of wood with a proportion of heartwood, water-soluble extractives in the heartwood are transported along with water to the wood's surface and are deposited as solids when water evaporates. As a result, the wood's surface is contaminated with extractives, which in turn affects the wood adhesion or bond strength and leads to low strength glue bonds. Hse and Kuo, (1988) further listed the different gluing interference mechanisms that can be caused by high level of extractives in heartwood:

- Heavy deposits of extractives on the gluing surface may block the reaction sites, thus preventing the anchoring of adhesives.
- Chemical incompatibility between the extractives and adhesives may result in inferior bonds.
- Extractives influence the wettability and polarity of the wood surface so that the wettability-permeability relationship of adhesive is changed.
- Extractives affect the curing and setting characteristics of adhesives.
- Oxidation of extractives tends to increase the acidity of wood and promote degradation.

2.4.3. Sapwood

Sapwood, which in the living tree provides the path for the flow of sap, remains permeable to liquids even after the timber has been seasoned (FAO, 1986). The permeability of sapwood provides pathways for the adhesives to penetrate and form strong adhesive bonds.

To test the influence of sapwood and heartwood on bonding strength, Kaygin and Tankut (2008) conducted an experimental study to determine the effects of heartwood and sapwood of scotch pine on bonding strengths, under different environmental conditions (control, submerged in cold water, boiling water and sea water) and three adhesives (PUR, epoxy, RF). The results showed that all sapwood specimens had a better bonding strength (see Figure 2-6), when compared to those of heartwood, irrespective of the adhesive applied or environmental condition exposed to. The lower bonding strength in heartwood specimens was related to the extractive substances it contained. Kaygin and Tankut (2008), concluded that the excessive extractives in the heartwood affected the pH level, which in turn influenced the adhesive hardening and therefore, reducing the bonding strengths.

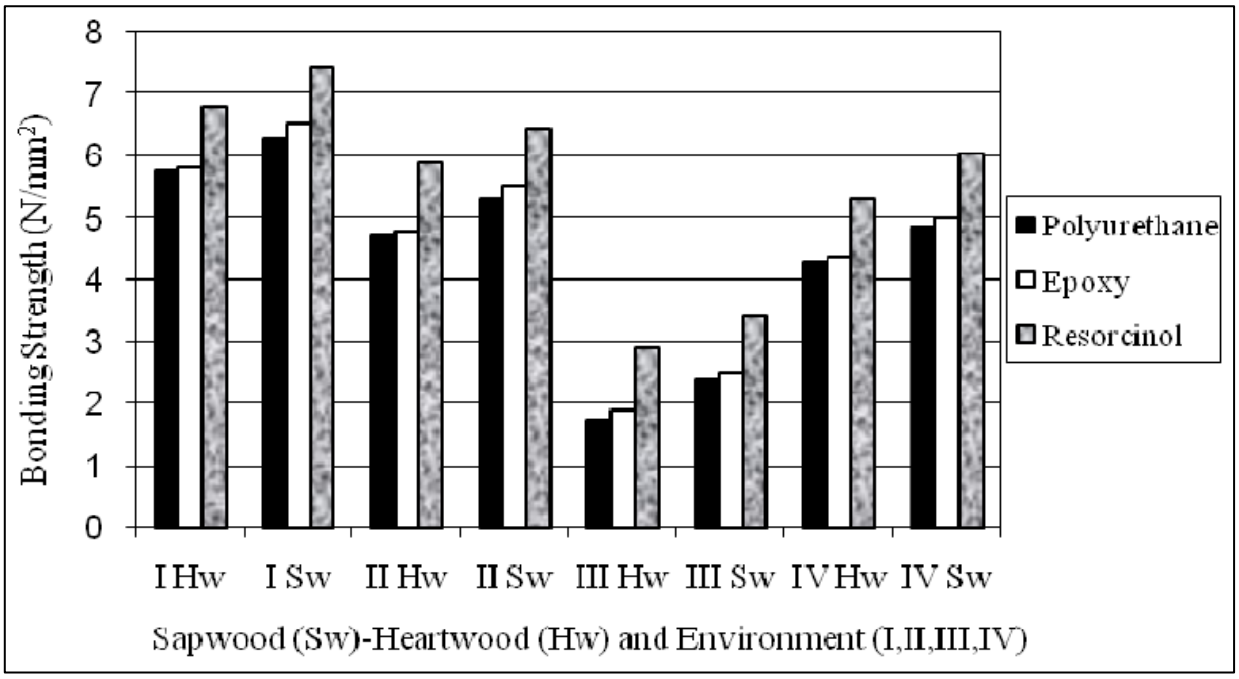


Figure 2-6: Bonding strength of Scotch pine wood according to the environment, sapwood-heartwood, and adhesive type (Kaygin and Tankut, 2008).

2.4.4. Moisture content

The presence of water in wood can influence the penetration of waterborne adhesives in two ways. When adhesives are applied on wood with low moisture content levels, the wood will draw the solvent (water), and polymer, more readily into the wood substrate (Kamke and Lee, 2007). As the water (water from the adhesive absorbed by the wood) is preferentially adsorbed by the dry cell wall, the effective solids content of the adhesive increases, leading to accelerated coalescence of the polymer and increased viscosity (Kamke and Lee, 2007). This may inhibit the penetration of adhesive (Honka, 2017) and cause bond lines of low strength as the adhesive coalesced polymers remain on the wood surface.

On the other hand, when waterborne adhesives, such 1C-PUR are applied on wood with high moisture content, excessive penetration and flow of the adhesive can occur (Sterley, 2012). This is due to the fact that 1C-PUR adhesives are moisture curing adhesive systems, which use moisture contained in the wood as the second component to react with and initiate fast curing (Pröller, 2017).

Gruver and Brown (2006) observed little penetration of pMDI adhesive at 0% moisture content, while extensive adhesive penetration was observed at 5% and 12% moisture content levels. The authors concluded that, as moisture content increased, shear strength (see Figure 2-7) and wood failure increased, due to greater/deeper penetration.

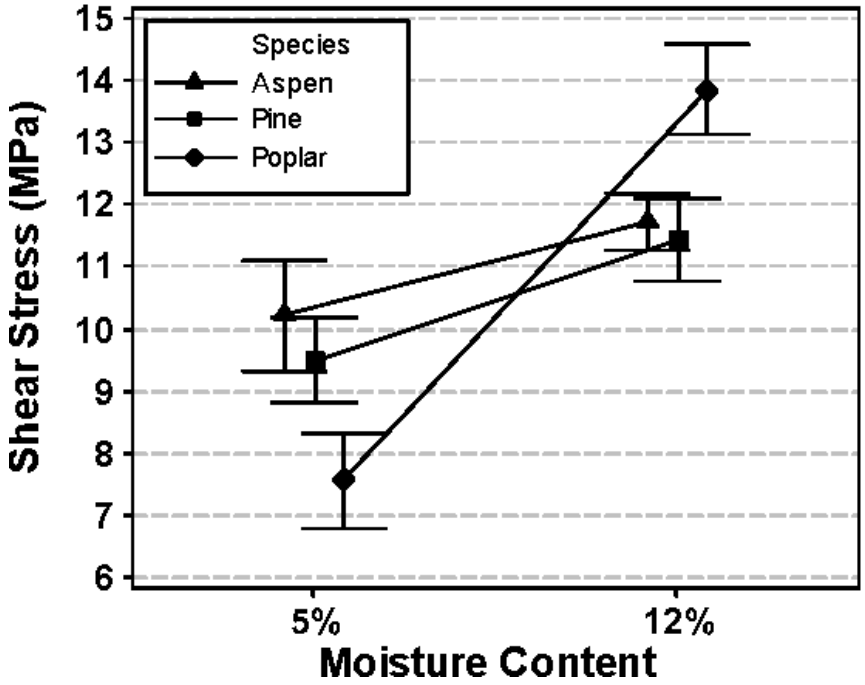


Figure 2-7: Shear strength results based on different moisture content levels (Gruver and Brown, 2006).

2.4.5. Wood surface

The presence of non-polar substances (e.g. dust particles, migrated extractives to the surface during drying) on the wood surface tend to reduce surface energy and retard wetting by aqueous adhesive systems (Kamke and Lee, 2007). In cases were wood has been treated with preservatives, the presence of wood preservative deposits (e.g. Cu, As, Cr) alters the wood surface properties causing the surface to become rougher, hydrophobic, acidic, reduce surface energy, increase contact angle and unevenness on the surface. The deposition of these preservatives may also cause blockages on the cell lumens and as a result, introduce stress concentrations when the adhesive has cured. However, planing is usually the best solution in improving surface properties amongst other techniques such as solvent application. Planing not only removes extractives contaminating the wood surface but also exposes a fresh and highly polar surface to which adhesives bond most efficiently (Hse and Kuo, 1988). How et al. (2017) also reported that freshly planed wood has a larger amount of opened lumen for the adhesive to fill, thus providing stronger adhesion.

2.4.6. Adhesive spread rate

Considering that the variability of the physical properties across a group of species can be large, it is necessary to identify the specific spread rate according to wood species in each fabrication attempt (How *et al.*, 2017). The physical properties of wood that can affect and dictate the amount of adhesive applied include porosity of wood, density, cellular structure, softwood/hardwood and moisture content. For instance, (Sterley, 2012) stated that 1C PUR adhesives are able to penetrate the cell structure of wet softwoods (high moisture content) up to twice as deep as compared to dry material. Therefore, due to the high adsorption, an increase of adhesive spread rate might be required.

2.4.7. Pressure

Pressure enhances wetting by forcing liquid adhesive to flow over the surfaces, displace air blockages and penetrate to the sound wood (Frihart and Hunt, 2010). Sikora, McPolin and Harte (2016), Wang *et al.* (2018), Vick and Okkonen (2000), found high bonding pressures to substantially improve the durability of adhesive bonds, as the adhesive penetrates deeper into the cellular structure and form mechanical interlocking. The greater penetration promotes a greater distribution of stress between the wood substrates when placed under load (Kamke and Lee, 2007). However, greater depth of penetration, with a fixed amount of polymer, may reduce the concentration of polymer in the bond line (Kamke and Lee, 2007) if the wood is too porous or has large lumen openings. This excessive penetration may leave insufficient adhesive on the bond line and lead to a starved bond line. On the other hand a lack of pressure can also impair the quality of the bond line due to insufficient penetration resulting in a thick glue line with CO_2 induced cavities (Sterley, 2012).

2.4.8. Molecular weight distribution

Molecular weight distribution of adhesive systems will impact their ability for cell-wall penetration (Kamke and Lee, 2007). The use of adhesives with low MW components has the potential for deeper penetration than that with high MW (Kamke and Lee, 2007). Adhesives that exhibit a lower molecular weight infiltrate or diffuse into the cell wall before curing. This is because of the low viscosity, which allows better flow and wets more of the surface. On the other hand, adhesives with higher molecular weight (e.g. 1C-PUR) often fail to penetrate the cell wall. They also tend to dry out quickly and have little penetration due to the high viscosity.

2.4.9. Adhesive type

Based on their chemistry, structure properties and their interaction with wood, adhesives can be mainly grouped into two groups namely: in-situ polymerized (e.g. UF, MF, MUF, PF, PRF, pMDI etc.) and pre-polymerized (e.g. PUR, PVAc, EPI etc.) (Frihart, 2009). *In-situ polymerized* adhesives (UF, MF, MUF, PF, PRF, pMDI), are highly cross-linked polymers (usually when cured) with a rigid backbone and are mainly thermoset. Generally, in situ *polymerized* adhesives exhibit a lower molecular weight than *pre-polymerized* adhesives, which causes it to infiltrate or diffuse into the cell wall before curing. On the other hand, *pre-polymerized* adhesives (PUR, PVAc, EPI, proteins, and mastics) are polymers with a flexible backbone, limited cross-linking, and usually have a higher molecular weight, which prevents it to penetrate the cell wall. They develop adhesive strength by losing water and/or by cross-linking the flexible polymers (Hunt *et al.*, 2019). These two groups of adhesives also differ significantly in their ability to distribute moisture-induced stress in an adhesive bond resulting in different failure mechanisms (Sikora, McPolin and Harte, 2016). For instance, when in-situ polymer bond lines are exposed to frequent wetting, they may fail as they do not have the ability to respond to moisture induced dimensional changes, such as swells in wood, which may potentially create large stresses at the interface (Frihart, 2009).

2.5. Wood-adhesive bond testing methods

2.5.1. Standards for wood-adhesive testing

Engineered wood products with several layers bonded together are controlled and regulated by standards, which ensure that the bonded products prescribe and meet the requirements in terms of strength, ability to

withstand mechanical load stresses and extreme climatic conditions (e.g. moisture changes, high temperatures).

Over the years, several standards have been developed for the assessment and evaluation of the strength and durability of interfacial adhesive bonds between laminates. In the US, ASTM standards, such as D905, D906, D2559, D7247 (Hunt *et al.*, 2019), ANSI standard for Glulam (ANSI 405) and ANSI/APA PRG 320 for CLT, (2012), are commonly used for testing the strength and integrity of bond lines. Europe mostly uses the CEN standards for bond line testing, which include EN 14080, 16351, 314, 391 and 392. In South Africa, SANS standards, such as 10183-4-2, 6044 and 1460, are commonly used for bond line assessment. Most of these bond-testing standards/methods more or less follow the same testing procedure, where the bonded products are certified by achieving a specific load (shear test), a certain wood failure percentage and the ability to withstand or resist delamination when exposed to accelerated harsh climatic conditions. These standardised procedures and test methods are essential for both quality control and certification (Betti *et al.*, 2016).

For instance, ASTM D 905 (2003) and EN 392 (1995) standards follow a similar testing procedure, where a self-aligning shearing force is applied via a cylindrical bearing at the end-grain, with a stress field uniform in the width direction. The difference in comparison between the two standards, is the preparation of test blocks (see Figure 2-8d and Figure 2-9). However, difficulties may arise with the ASTM D905 (2008) standard during the preparation of test specimens, as cutting errors (Figure 2-8 e, f) may arise and affect the shear strength values (Derikvand and Pangh, 2016). Nonetheless, by the cutting the specimens in a staggered shape as in ASTM D905 (2008), this exposes the glueline much better than EN 392 (1995) (Figure 2-9), as the load is directly loaded on the glueline. Furthermore, by cutting the test samples in a staggered shape, the effect of slope of grain is minimized.

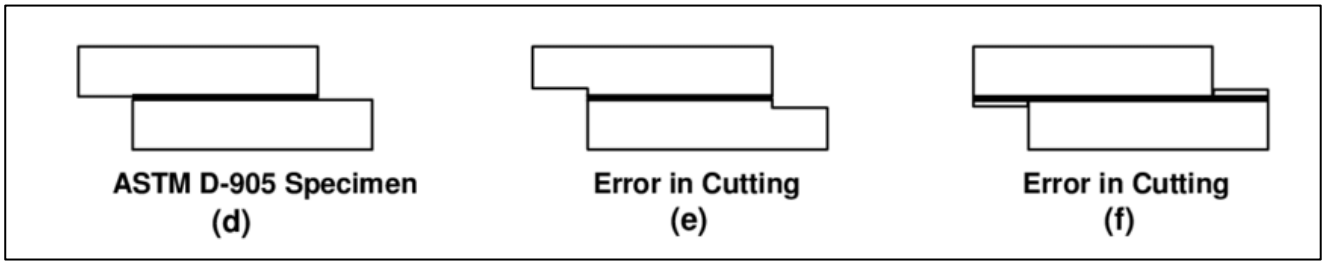


Figure 2-8: ASTM D905 (2003) test specimen configuration (d, e, f).

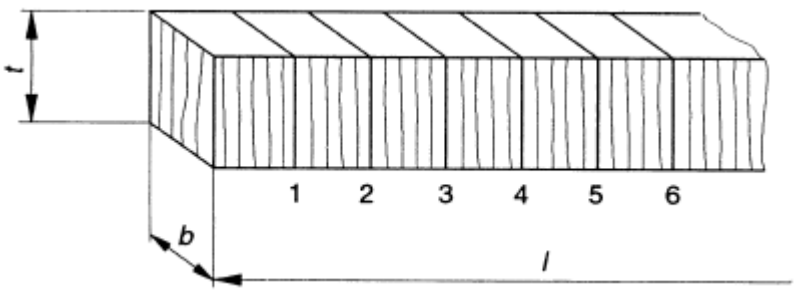


Figure 2-9: EN 392 (1995) test specimen configuration.

In the case of delamination standards, most are quite similar are carried out by submerging test specimens in water and applying pressures and drying them rapidly and measuring the cracks in the bond line caused by

the rapid drying. Delamination standards often differ in parameters, such as the impregnating-drying cycle's duration (depending on the level of exposure), drying temperature and specimen size.

2.5.2. Shear strength, resistance to delamination and wood failure percentage

Shear test

Shear tests serve as a useful criterion for the estimation of mechanical compatibility between wood and the adhesive. In theory, shear tests expose only the glue line of the specimen to shear so in this way the strength of the bond is tested (Weidman, 2015). The advantage with shear tests is that they are relatively easy to perform and provide quick objective results, but there are several shortcomings that should be noted. Steiger and Richter (2009) highlighted that in most shear strength test methods, the resulting stress in the bond line is not pure shear but rather a combination of shear and normal stresses. Furthermore, certain weaknesses have been observed in shear testing methods, such as the non-uniform shear stress distribution and rolling shear in cross laminated timber. Several researchers have also mentioned/listed other factors that may affect the measured shear strength such as sample geometry, wood strength, surface roughness of wood, wood defects, adhesive formulation, wood preservatives, shear tool design, sapwood/heartwood, wood density, growth ring orientation and the rate of loading (Okkonen and River, 1989; ASTM D 905, 2003; Burdurlu *et al.*, 2006; Steiger and Richter, 2009; Wang *et al.*, 2018).

In an attempt to investigate the extent of some of the factors, Gaspar, Cruz and Gomes, (2008) tested the effects of sample size and wood preservatives on shear strength and wood failure percentage. Regarding size effect, the shear strength results of both wood and gluelines was found to decrease with the increase of the specimen dimensions (Table 2-1), whether treated or not.

Table 2-1: Results of block-shear specimen tests (Gaspar, Cruz and Gomes, 2008).

Glulam type	Specimen dimensions (mm x mm)	Glueline shear strength			Average WFP %	Wood shear strength			Correlation*
		N^{er} of values	Average (N/mm^2)	Standard deviation (N/mm^2)		N^{er} of values	Average (N/mm^2)	Standard deviation (N/mm^2)	
Not treated maritime pine	20 x 20	104	15,9	22	89	133	16,5	2,5	0,41
	35 x 35	49	14,4	1,7	91	60	15,9	2,3	0,21
	50 x 50	41	12,7	1,3	86	64	13,3	2,1	0,31
Spruce	20 x 20	106	11,3	1,8	87	163	12,4	2,6	0,3
	35 x 35	60	10,8	1,2	88	99	11,8	2,4	0,09
	50 x 50	43	9,8	0,9	90	68	10,8	2	0,23
Treated maritime pine	20 x 20	72	16,5	2,3	78	103	17,5	2,1	0,12
	35 x 35	36	14,6	2,3	73	79	16,8	1,9	0,1
	50 x 50	30	13,2	2,1	72	51	16,4	14	0,03
*-Correlation coefficient of the regression line between glue line shear strength and wood shear strength									

Delamination test

Delamination tests are designed to assess the integrity of gluelines between laminates by imposing fast changes in moisture content inducing significant shrinkage and swelling response in a relatively short period of time (Tascioglu, Goodell and Lopez-Anido, 2003). The shrinkage and swelling in the laminates caused by

the fast changes in moisture content, affects the bond integrity and results in openings on the gluelines. For bond lines to be approved, they have to withstand the expansion and shrinking that occurs when the wood picks up and loses moisture content, as well as the thermal expansion and contraction of the wood (Frihart, 2003). This process allows the determination of whether the gluelines can withstand different climatic conditions over the useful life of the product. However, delamination test results are known to be inaccurate and very subjective, as they are affected by several factors, including size of specimen, wood species, adhesive formulation, number of cycles, wood density, wood defects.

According to a study conducted by Betti *et al.*, (2016), the delamination test results showed that the size of the specimen is a crucial factor in determining the outcomes of delamination tests. Dugmore (2018), also found that due to the large specimen size (100 x 100 mm) as prescribed by EN 16351 (2015), excessively high stresses formed by shrinkage and swelling are created, leading to increased delamination for CLT specimens. Knorz, Torno and van de Kuilen, (2017), highlighted that stresses caused by a moisture gradient within specimens during impregnation and drying in the delamination test vary with the specimen shape and therefore, influence delamination results.

According to Sikora, McPolin and Harte (2016) the prediction of bond line strength using both shear and delamination test methods, is highly dependent on the specimen type used and the adhesive properties. To limit or minimize effect of these factors, wood failure percentage is often recorded for further bond analysis as it provides information on whether the strength is in the wood or the adhesive bond.

Wood Failure Percentage

As much as shear and delamination tests are the two main methods prescribed by standards of laminated wood as means of evaluating the bond integrity and strength of bond lines, the susceptibility of these test methods to wood properties, adhesive formulation and test tools, often makes it difficult to record pure/true test results. As a result, wood failure percentage is often recorded as means of further assessing the wood-adhesive bond. Steiger, Arnold and Risi (2014) describes wood failure percentage as a method that indicates, which of the materials (wood or adhesive) is weaker in terms of the ratio of areas that failed within the glue line (cohesive failure) or in the wood-adhesive interface (adhesion failure), and those failed in the wood itself. Sikora, McPolin and Harte (2016) argue though that, as much as wood failure provides information on whether the superior strength is in the timber or the bond, it lacks information on the failure behaviour. Furthermore, the splitting process for determining the WFP in delamination tests, which is done by using a hammer and chisel to split the glueline, is highly criticised by various researchers, due to its subjectivity when determining the percentage estimation.

Because of these limitations in the methodologies used for assessing adhesive bonds performance, it is generally accepted that no single test procedure can provide all of the information to definitively measure bonding quality and strength (Sikora, McPolin and Harte, 2016).

2.6. Performance of glued treated wood

Although wood preservatives offer a solution to the deterioration of mass timber structures caused by moisture pickup, insects and fungal growth, some studies have reported that the inorganic material deposited by the preservatives may interfere with the formation or development of durable bonds. The presence of preservatives

may influence the formation of bonds in one of the following ways: alter the adhesive pH and cure rate, increase surface hydrophobicity and roughness, increase oxygen/carbon ratio and reduce carbon (Tascioglu, 2007); alter surface pH (Maldas and Kamdem, 1998); cause high moisture content in the wood, inhibit the condensation reaction, delay water removal from the glueline, prevent adhesive from wetting the wood surface (Raknes, 1964); and reduce lumen opening (preservative accumulation on the lumen surface).

Tascioglu, Goodell and Lopez-Anido (2003) reported that the insoluble metal oxides contained in CCA/CCB preservative solutions, tie up aromatic hydrocarbon functional groups reducing hydrogen bonding and/or perhaps covalent bonding opportunities between the adhesive and lignocellulosic wood content. Ostmeyer et al. (1989) showed that all three components of CCA preservative formed chemical bonds with the aromatic ring of lignin and the carbonyl groups present in wood. Maldas and Kamdem (1998) similarly reported that the metal oxides deposited in the cell lumen, may reduce the opportunities for stable chemical bonding between the wood components and adhesives, as the adhesive might fail to wet the lumen surfaces.

Vick (1994) used scanning electron microscopy (SEM) coupled with energy-dispersive X-ray analysis (EDXA) to show that the inside of cell lumen of CCA-treated southern pine was completely covered with a mixture of copper, chrome and arsenic. However, the microscopic analyses also showed that the deposits were not large enough to block openings of a bordered pit aperture (see Figure 2-10), which must remain open in order allow the adhesive to flow into the lumen and form mechanical interlocking between the adhesive and wood substrates.

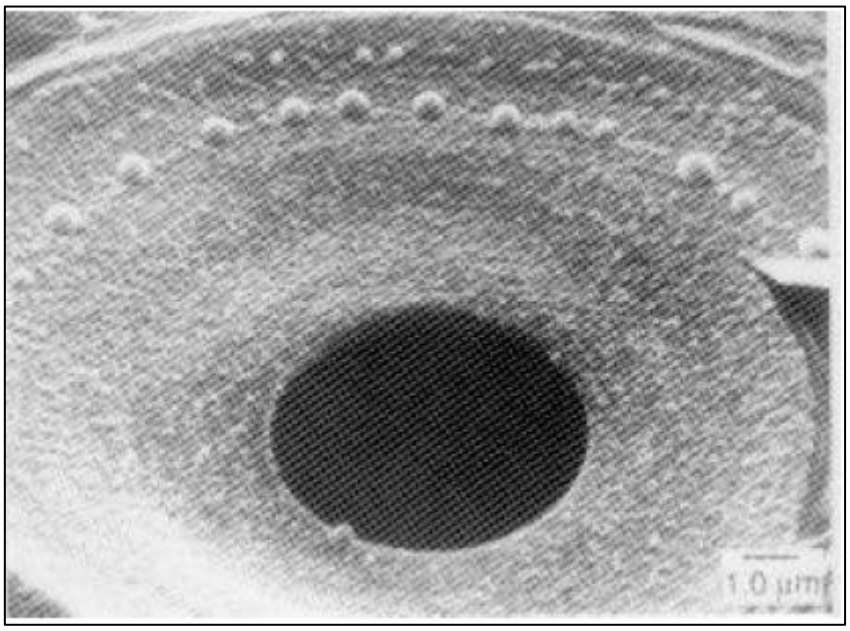


Figure 2-10: Bordered pit aperture showing a relative size of metal deposits to the opening through which adhesive flows (Vick, 1994).

Shear strength:

In a study conducted by Özçifçi (2006) to evaluate the effect of five wood preservatives (borax, boric acid, CCA, mixture of borax + boric acid and mixture of di-ammonium phosphate + borax + boric acid) on the bond strength of phenol-formaldehyde (PF) and melamine-formaldehyde (MF) adhesives, the results showed lower shear strengths on wood samples pre-treated with CCA than those of borate-based preservatives. For CCA,

this cause was attributed to CCA solutions having a high degree of acidity and high extent of retention and thus, leading to the deterioration of the wood surface. It was further found in the study that, as the extent of retention of CCA or any impregnating preservative increases, the adhesion between the adhesive and the wood often decreases.

In a similar study, Zhang *et al.* (1997) reported a 20% shear strength loss in CCA treated southern pine, bonded with resorcinol formaldehyde (RF) adhesive. However, the wood failure percentage did not show any significant difference.

Ozdemir, Temiz and Aydin (2015) reported that scots pine (*Pinus sylvestris*) specimens impregnated with aqueous solutions of 1% boric acid and 2% copper azole (Tanalith E), provided increased adhesion strength (in accordance to ASTM D 4541) when compared to untreated samples (Figure 2-11). This increased adhesion strength of boric acid treated specimens was attributed to higher mechanical interlocking mechanism of adhesion caused by increased surface roughness of boric acid-treated wood. However, for wood specimens impregnated with aqueous solutions of 2% CCA and Immersol aqua, a decreased adhesion strength (Figure 2-11) was reported. The low adhesion strength in CCA was attributed to poor wetting properties caused by the deposition of CCA oxides, while the poor adhesion strength in organic borne Immersol aqua was due to the decreased bonding capacity of coatings and wood.

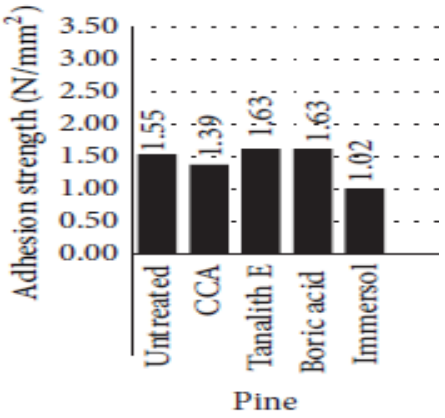


Figure 2-11: Adhesion strength of untreated and treated samples (Ozdemir, Temiz and Aydin, 2015).

Vick, De Groot and Youngquist (1990) investigated the compatibility of 13 non-acidic waterborne preservatives with phenol-formaldehyde (PF) adhesive. All three borate-based preservatives (ammoniacal copper borate, ammoniacal pentaborate, disodium octaborate tetrahydrate) included in the study, caused poor bonding even at the lowest retention levels.

Cameron and Pizzi (1985) assessed the effect of overtreatment on the strength of PRF gluelines in *Pinus patula*. Contrary to several findings, the authors found that the shear strength is unaffected by increased levels of CCA retention, although the wood failure percentage is decreased. Interestingly, out of all the retention levels tested (0, 16, 20, 32 kg/m^3), the 32kg/m^3 recorded the highest shear strength (11.96 MPa) at a PRF spread rate of 200g/m^2. Cameron and Pizzi (1985) explained that once chromium fixes onto the wood surface, chromium forms a strong, stable and irreversible complex with phenolic adhesive resulting in higher surface wetting, while the low wood failure could be due to lack of adsorption of the adhesive on to the already heavily

coated wood fibres. Vick (1994) also highlighted that chromium metallic ions (Cr^{+3}) in CCA, forms a stable complex with resorcinol-formaldehyde and phenol-formaldehyde adhesives.

Lim, Tripathi and Tang (2020) also reported interesting findings when testing the effect of micronized copper azole type C treatment (at two retention levels 1.0 and 2.4 kg/m^3) on CLT manufactured from southern yellow pine with three adhesive systems (MF, RF and PUR). The low and high retentions of MCA-C treatment on MF resin (see Figure 2-12), significantly reduced the shear strength in comparison to untreated blocks. However, for the RF and PUR adhesives (Figure 2-12), only the low retentions showed a significant reduction in shear strength, while the untreated and high retention treatments exhibited higher shear strengths.

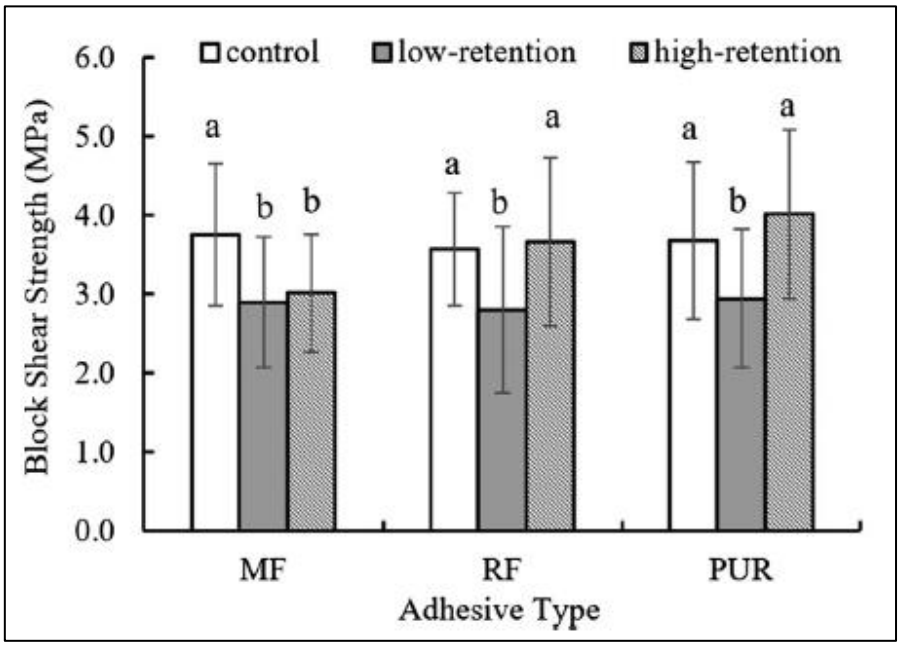


Figure 2-12: Mean block shear strength of CLT configurations by different adhesive types (bars with different letters are significant) (Lim, Tripathi and Tang, 2020).

Gaspar *et al.*, (2010) conducted a study to evaluate the performance of glued laminated timber of maritime pine treated with a copper azole preservative at different retention levels. The shear strength of the glue lines met the EN 386 (2001) requirements and some of the gluelines of preservative treated wood were slightly higher than for untreated test specimens. The improved adhesion strength on copper azole treated wood was also reported by (Ozdemir *et al*., 2015). However, delamination (in Figure 2-13) increased with the increase of the preservative retention rate. Gaspar *et al.*, (2010) concluded that wood preservatives have a negative influence on the glue line behaviour as delamination increases with higher retention rates and as such, recommended that the preservative retention rates for glued laminated timber should be as small as possible.

	Wood treatment level		
	Z	L	H
Number of specimens	14	15	13
Average delamination (%)	0.66	0.91	2.38

Figure 2-13: Results of the average delamination for each wood treatment level/retention (Z=untreated, L= low 7.6 kg/m^3 and H= high 19.1 kg/m^3.

Gaspar et al. (2008) also tested the effect of copper azole (*Tanalith E* 3492) and sample size on shear strength and wood failure percentage of maritime pine. The shear strength results revealed that treated maritime pine produced stronger gluelines than untreated maritime pine. However, the presence of preservatives on maritime pine seemed to have a negative effect on wood failure, when compared to untreated maritime pine.

Delamination:

In a delamination test of southern pine treated with CCA, Zhang et al. (1997) highlighted that CCA treatments improved resorcinol formaldehyde (RF) adhesive wettability, as expressed by the lower contact angle of RF on the CCA treated than on the untreated southern pine surface.

To understand the effect of preservatives and their retention levels on bond durability, Tascioglu (2002) assessed the effects of increasing CCA (type C) retention levels on delamination of PRF bond lines in wood/wood (southern yellow pine) and wood/FRP (fiber reinforced polymer). The lamellas were pre-treated with a full cell process individually and the ASTM D 2559 three cycle aging test was carried out on the test blocks. Retention levels from 4.65 to 10.3 kg/m^3 (see Figure 2-14) caused an increased delamination (from 0 to 13%) for wood/wood (SYP) interfaces, however, a decline in delamination for retention levels from (23.9 to 51.4 kg/m^3) was observed. After conducting a surface energy analysis (using a SEM microscope), the bimodal delamination response to CCA retention levels was explained by the fact that the total surface energy of CCA treated southern pine increases with increasing retention (Tascioglu, 2002). The increased surface energy was attributed to the chemical modification of the wood surface by the high surface energy metallic salts (Tascioglu, 2002). Nonetheless, Tascioglu (2002) concluded that laminates pre-treated with CCA negatively interfered with the bond durability.

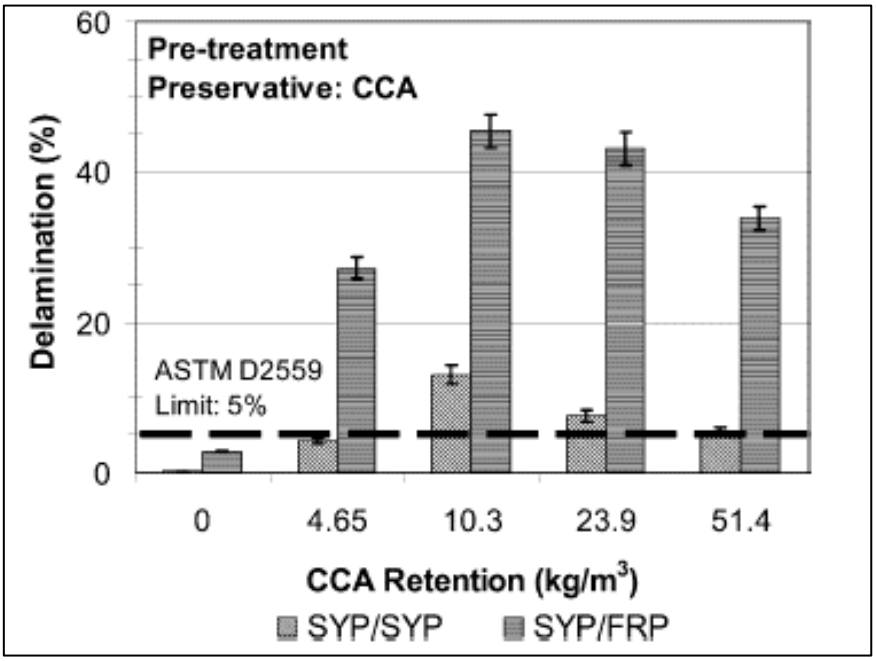


Figure 2-14: Effects of increased CCA retention on delamination of PRF bond lines (allowable maximum delamination = 5%) (Tascioglu, 2002).

Lorenz and Frihart (2006) examined the effect of CCA, ACQ and CA-B preservatives on the bond durability of southern yellow pine bonded with PRF adhesive. Interestingly, the delamination results showed that CCA

did not interfere with bond formation as the delamination results were comparable to untreated sample results as shown in Table 2-2. The authors also found (using a Differential scanning calorimetry) ACQ and CA-B preservatives to accelerate the reaction of the PRF adhesive, which lessened the penetration of the adhesive into the wood.

Table 2-2: Delamination of southern yellow pine bonded with PRF.

Wood treatment	Primer concentration	Delamination (%)
None	None	1.7 to 3.0
CCA	None	0.4 to 1.2
CA-B	None	2.0 to 8.9
ACQ	None	6.5 to 15

Pizzi (1979), cited in Maldas and Kamdem (1998), investigated the effect of CCA treatment on the curing behaviour of RF and PF adhesives. The bivalent copper Cu^{2+} ions in CCA were found to have an accelerating effect on the curing rate of adhesive, reducing gel time of the adhesive, while the trivalent chrome Cr^{3+} ions had a retarding effect by increasing the normal gel time. The presence of CCA deposits suggested that they might cause high delamination bonds when exposed to severe cyclic aging tests. Contrary, Vick and Christiansen (1993) conducted a similar experimental study, to analyse the cure and reaction of phenolic adhesive in CCA-treated to untreated southern pine using a differential scanning calorimetry. Results showed that once CCA had fixated within the wood, the CCA preservative metallic ions did not interfere with the normal curing reaction of PF adhesive.

Chapter 3 : Materials and Methods

The experimental work of this project was aimed at evaluating the effects of waterborne preservatives on the bond line performance of 1C-PUR adhesive. In addition, wood properties (density and sapwood/heartwood ratio) were also added as part of the experimental study. Common destructive adhesive testing methods including resistance to delamination, shear strength and wood failure percentage were adopted for the experiment. A statistical analyses test was carried out to analyse the data and determine whether the treatments (CCA and DOT) and wood factors (sapwood/heartwood ratio and density) have a significant effect on the bond strength and durability of 1C-PUR adhesive. Timber from the most planted softwood in South Africa, *Pinus patula*, was used.

The research procedures shown in Figure 3-1 are discussed in more detail in the following sections.

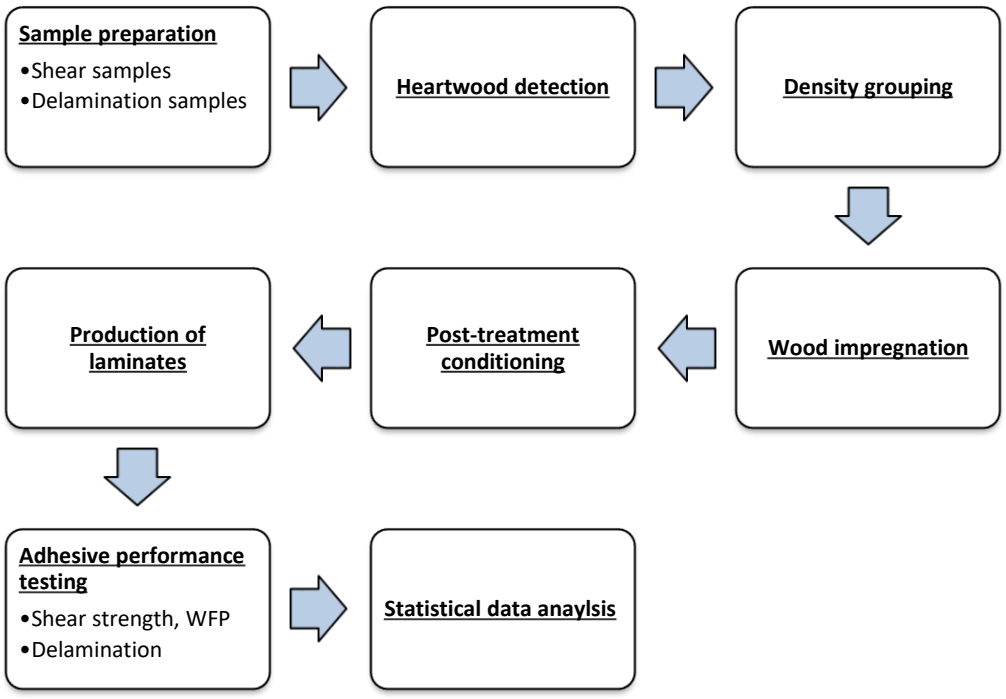


Figure 3-1: Experimental procedure.

3.1. Materials

3.1.1. Wood

Grade S5 structural *Pinus patula* timber of 3.0 m x 38 mm x 152 mm (untreated) was purchased from a local supplier in the Western Cape Province. The timber had been kiln dried to a moisture content of 12±3% prior to being delivered. Upon delivery, the timber was stored under room conditions for 31 days.

3.1.2. Adhesive

A one component polyurethane (LOCTITE HB S409 PURBOND) cold setting structural adhesive system was used in this study. The 1C-PUR adhesive was formulated for the manufacture of engineered wood products and was classified as a type 1 adhesive. The LOCTITE HB S409 PURBOND adhesive complied with the

requirements of SANS 10183-2 (2014), EN 15425 (2008) and EN 301 (2006) standards. The specifications provided by the manufacturer were as follows: assembly time 40 minutes, minimum press time/curing time 100 minutes at 20°C and 65% RH, pressing pressure 0.6 – 1.0 N/mm^2, spread rate of 140 – 180 g/m^2, and wood moisture content of not less than 8%.

3.1.3. Wood preservative chemicals

The following wood preservative chemicals were used for treatment:

- A 60% commercial concentrate of CCA type C *(tradename: Permacure Oxide Liquor)* was supplied by Dolphin Bay Chemicals in the form of a homogeneous liquid that was readily miscible with water. The preservative had a pH of 1.68 and a density of 1.78 kg/dm^3.

- DOT ($Na_2B_8O_{13}.4H_2O$)*(tradename: Rentobor)* was supplied by Dolphin Bay Chemicals, in the form of powder and complied with the requirements of SANS 871 (2009) Type I. It contained 939g/kg boric acid equivalent.

3.2. Methods

3.2.1. Experimental design

As shown in Table 3-1, the wood preservatives (CCA and DOT) were used at different concentration levels in order to achieve retention levels suitable for different environmental exposure levels (hazard classes of H2, H3 and H4). Wood factors including density (low and high) and sapwood/heartwood percentage (Table 3-1) were also added as part of the experiment, as several studies (Tascioglu, 2007) have reported on their influence on bond strength and durability.

Table 3-1: Experimental factors.

	Factors	Unit	Factor levels	
			Low level	High level
Treatments	CCA	(w/v)	2%	4%
	DOT	(w/v)	1.67%	3.33%
	Untreated	-	-	-
Wood factors	Wood type	%	Heartwood>35%	Sapwood 100%
	Density	kg/m^3	376 - 462	473 - 557

The combination of these experimental factors, along with their factor levels led to the creation of 8 (2x2x2) combinations per preservative and 4 (2x2) combinations for the control (untreated). In total, the experimental study had 20 experimental groups (see Table 3-2, 3-3 and 3-4):

Table 3-2: Experimental design of CCA preservative groups.

Group	1	2	3	4	5	6	7	8
CCA	2%	4%	2%	4%	2%	4%	2%	4%
Wood type	Sapwood	Sapwood	Sapwood	Sapwood	Heartwood	Heartwood	Heartwood	Heartwood
Density	Low	Low	High	High	Low	Low	High	High

Table 3-3: Experimental design of DOT preservative groups.

Group	9	10	11	12	13	14	15	16
DOT	1.67%	3.3%	1.67%	3.3%	1.67%	3.3%	1.67%	3.3%
Wood type	Sapwood	Sapwood	Sapwood	Sapwood	Heartwood	Heartwood	Heartwood	Heartwood
Density	Low	Low	High	High	Low	Low	High	High

Table 3-4: Experimental design for control (untreated) groups.

Group (control)	17	18	19	20
Wood type	Sapwood	Sapwood	Heartwood	Heartwood
Density	Low	High	Low	High

3.2.2. Sample preparation

The 3 m long planks of *Pinus patula* were cut and planed into 20 mm (thickness) x 51 mm (width) x 310 mm (length) dimensions for shear strength samples and 32 mm (thickness) x 110 mm (width) x 500 mm (length) for delamination samples in accordance to SANS 10183-4-2 (2009). The SANS 10183-4-2 (2009) standard recommends the width of the lamella to be 150 mm for delamination, however, due to the limiting diameter size of the treating cylinder, the width was reduced to 110 mm. In CLT delamination testing, the reduction in width of the samples might result in lower delamination as Betti *et al.* (2016); Knorz, Torno and van de Kuilen (2017) and Dugmore (2018) found that large specimen size lead to increased delamination since excessively high stresses are formed by shrinkage and swelling. However, since laminations for this experiment was done with parallel grain direction, this reduction width will likely have a limited effect on results. Also, results will be conservative since the smaller sizes will likely increase delamination.

The wood samples were then stored in a conditioning room (65% ± 5% RH, 20º ± 2ºC) for a period of 7 days in order to achieve a uniform moisture content of approximately 12±3% in all samples.

3.2.3. Heartwood/sapwood percentage determination

Wood often contains varying proportions of sapwood and heartwood and to determine the amount or proportions of these wood tissues, the SANS 5999 (2004) method of detecting sapwood and heartwood was followed. A methyl orange indicator (ACS reagent, dye content 85 %) was used to distinguish between the sapwood and heartwood. The indicator was brushed on the cross section (both ends) of the wood samples

and the heartwood-sapwood boundaries were marked and estimated in percentages. The heartwood was coloured orange or red and sapwood remained pale yellow as shown in Figure 3-2. The wood samples were then separated into two groups: sapwood 100% and specimens were the heartwood percentage was higher than 35%. Ideally, samples with 100% heartwood would have been preferred but since it was almost unattainable to have the required number of samples with pure heartwood (as it would have required an enormous volume of timber), samples with a percentage above 35% of heartwood were selected and grouped under heartwood samples (35% heartwood limit allowed to have an equal number of samples between heartwood and sapwood). Take note that where reference is made in this thesis to heartwood samples it will mean the sample group where heartwood constituted more than 35% of the volume of each specimen.

This was followed by storing the samples in a conditioning room (65% ± 5% RH, 20º ± 2ºC) for 7 days in order to achieve a uniform moisture content of approximately 12±3% in all samples.

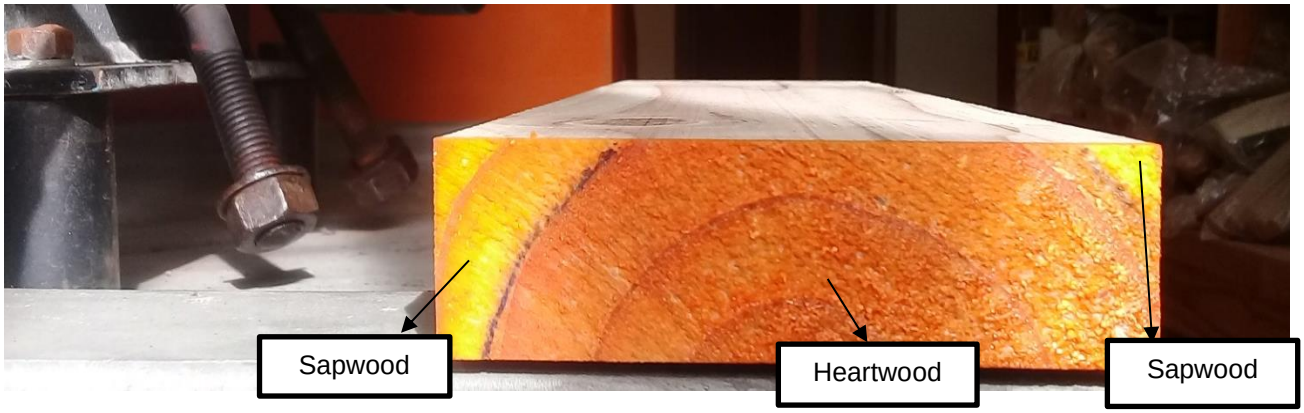


Figure 3-2: Heartwood and sapwood detection in specimens.

3.2.4. Density grouping/profiling

The wood samples were further separated into two density groups viz. a low and a high-density group (Table 3-5). The density of the wood samples was calculated through weighing each sample individually at approximately 12 ± 3% moisture content and dividing it by its measured volume.

Table 3-5: Density groups.

Density *(kg/m^3)*	
Low density	High density
376 - 462	473 - 557

Pre-treatment conditioning: after grouping the samples into two density groups, the samples were further conditioned for 7 days before wood treatment.

3.2.5. Wood impregnation process

The lamellas were pre-treated before gluing since the objective of this study was to evaluate the effect of treatment on bond quality.

The wood impregnation process was performed in a steel pressure cylinder, with a maximum pressure capacity of 640 kPa and volume capacity of 19.34 L (202.7 mm diameter x 600mm length). The wood impregnation process followed a Lowry empty-cell method of impregnation, which was in line with SANS 1288 (2016), SANS 10005 (2016), and ASTM D1413 (2007) standard requirements. However, certain modifications were made on the empty cell process due to the limitations of the equipment used. The pressure cylinder had no drainage outlet and as such, the impregnation process in this experiment excluded the final vacuum as prescribed.

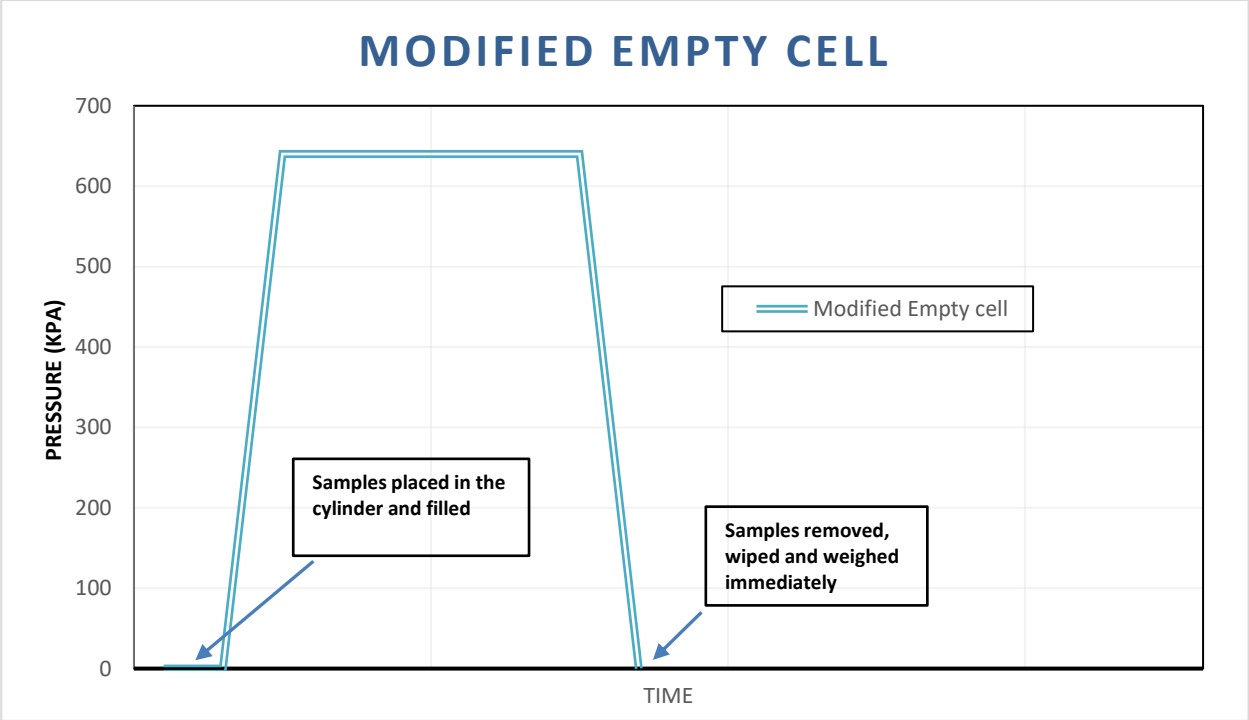


Figure 3-3: Modified empty cell treating cycle.

***Solution preparation**:* the samples were treated with water-soluble CCA and DOT preservatives. The aqueous solutions were prepared at different mass concentration levels shown in Table 3-6. The mass concentrations were derived and based on the average net retention requirements of SANS 1288 (2016) and SANS 10005 (2016) for softwoods and solution uptake capacity of samples. According to SANS 1288, when softwood sawn products are pressure treated with CCA preservative for an H2 hazard class, a minimum retention rate of 6kg/m^3 must be obtained. Similarly, for H4 hazard class a minimum retention rate of 12kg/m^3 must be reached. For borate-based compounds, a minimum retention rate of 5kg/m^3 must achieved for the H2 hazard class. The information on targeted retention, mass concentrations and solution strengths are presented in Table 3-6.

Table 3-6: Preservative solutions and targeted retention rates for CCA and DOT.

Preservative	Hazard class	Targeted retention (kg/m^3)[(a)]	Mass concentration (g/l)	Solution strength (w/v %)
CCA	H2	6 kg/m^3	20 g/l	2 %
	H4	12 kg/m^3	40 g/l	4 %
DOT	H2	5 kg/m^3	16.67 g b.a.e./l	1.67 %
	H2	10 kg/m^3	33.30 g b.a.e./l	3.33 %

[a]*Targeted retention as per SANS 10005 (2016) and SANS 1288 (2016) requirements*

***Treatment procedure**:* in order to determine the retention rate (*kg/m³*), each lamella/sample was pre-weighed *(T_1)* before treatment. After weighing, the wood samples were placed in the cylinder and a cylindrical metal weight was placed on top of the samples to avoid the movement of the samples during pressure application. The treating cylinder was then filled with an aqueous solution of CCA or DOT until wood samples were completely immersed. The treating cylinder door was then closed and pressure applied (pressure was supplied through a compressor connected to the treating cylinder) and increased to a maximum of 640 kPa and maintained for 30 minutes. When the pressure period had elapsed, pressure was then released. After completion of the pressure application, the wood samples were then removed from solution and lightly wiped off to remove excess solution on the surface. Each sample was then re-weighed (T_2) immediately (to the nearest 0.01g) in order to calculate retention rate using Equation 1. The aqueous solutions were not used to treat more than two charges.

$$Retention\ (kg/m^3) = \frac{G\ \times C}{V} \times 10\frac{kg}{m^3} \quad (1)$$

G: ($T_2 - T_1$) grams of treating solution absorbed by the wood sample (solution uptake)

C: solution concentration (%) or grams of preservative in 100g of the treating solution

V: volume of sample (cm^3)

Post-treatment conditioning: After re-weighing, the samples were placed on racks and exposed to open laboratory room conditions for 72 hours (ASTM D1413, 2007) to allow the release of the excessive preservative solution. The samples were then placed in a conditioning room (65% ± 5% RH, 20º ± 2ºC) for 21 days in order to allow the active ingredients to fixate into the wood and also lower the moisture content back to the prescribed range of 12% ± 3% (see Figure 3-4). After 21 days, the samples were monitored through successive weighing until they reached an equilibrium weight.

Figure 3-4: Delamination samples (left): A – 3.33% DOT, B – 1.67% DOT, C – 4% CCA, D – 2% CCA, E - Untreated; Shear samples (right): F – CCA treated and G – DOT treated samples.

3.2.6. Production of laminates

Moisture content: As per requirement of glued laminated product standards (EN 14080, 2013) and SANS 1460 (2015) laminates should be bonded when the moisture content is within the range of 12% ± 3%.

The moisture content of the samples was determined according to SANS 5984 (2004) oven dry method. Each group had additional samples, which were cut and reduced into 75 mm length blocks for moisture content determination.

Planing: To improve surface properties (e.g. contact angle, surface energy and penetration) and ensure strong and durable bond lines, the laminates were planed to get a fresh wood surface. In order to prevent the efflorescence of salts from wood preservatives, the planing time was not more than 8 hours before bonding. The delamination and shear lamellas were planed to a thickness of 30 and 19 mm, respectively, using a surface planer and a thickness planer. In most cases, the samples were still fully saturated or covered with the preservative even after planing, since a very thin layer was removed.

Adhesive application: Each lamella was first wiped with a clean cloth to remove dust particles (Dugmore, 2018). A 1C-PUR cold setting adhesive system was applied on one side of the tangential face of each lamella, using a spatula at a spread rate of 150g/m^2 at a room temperature of 20 ± 2 °C, as recommended by SANS 10183-2 (2014) and the adhesive manufacturer. Based on the recommended adhesive spread rate of 150g/m^2, the required amount of adhesive (g) per bond line was 2.37g and 8.25g for shear (310mm x 51mm) and delamination laminates (500mm x 110mm), respectively.

To achieve the required amount of adhesive (g) per bond line, the mass or amount of adhesive applied on each lamella was determined by tare and weight. The assembly time for all the samples was less than 15 minutes.

Pneumatic press: A pneumatic press system shown in Figure 3-5 was used to press the laminates at a pressure of 600 kPa. The pneumatic press system achieved the targeted pressure by pushing compressed air into hoses to force two rigid steel plates towards each other where the laminates were inserted between steel plates (Dugmore, 2018).

Figure 3-5: Delamination laminates under pressure in a pneumatic press system.
***Note:** portions of the assembly that extended or overlapped beyond the press plates were not used for testing purposes.*

The adhesive manufacturer recommended a press time or curing time of 100 minutes; however, the press time was extended to 120 minutes. This is because Tascioglu (2007) and Roffael (2016) reported that preservatives may delay hardening of the adhesive.

For the production of delamination laminates, six flat sawn lamellas were bonded in accordance to SANS 10183-4-2 (2009) and two flat sawn lamellas were bonded in accordance with ASTM D905 for shear test laminates.

After curing, the laminates were stored in a conditioning room (65% ± 5% RH, 20º ± 2ºC) for a period of 7 days.

3.2.7. Test blocks

Trimming and cutting: after the re-conditioning period had lapsed, the laminates were trimmed due to the foaming and swelling of 1C-PUR adhesive after curing. The shear laminates were trimmed and width reduced to 50mm, while the width of the delamination laminates was reduced to 105mm. Shear (50 x 44 mm) and delamination (75mm thick) test blocks were then cut from each laminate as shown in Figure 3-6 and Figure 3-7.

Shear strength test blocks:

Four test blocks were obtained from each laminate of shear test (see Figure 3-6).

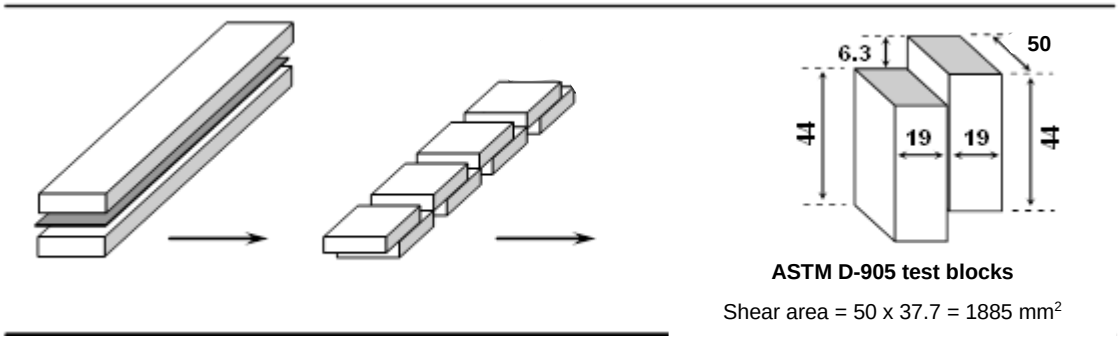


Figure 3-6: Shear strength test blocks.

Delamination test blocks:

From a full cross-section of each laminated member, four 75mm thick specimens (see Figure 3-7) were extracted by cutting perpendicular to the surface of the assembly (SANS 10183-4-2, 2009). No specimens were obtained closer to 50mm of each end of laminate.

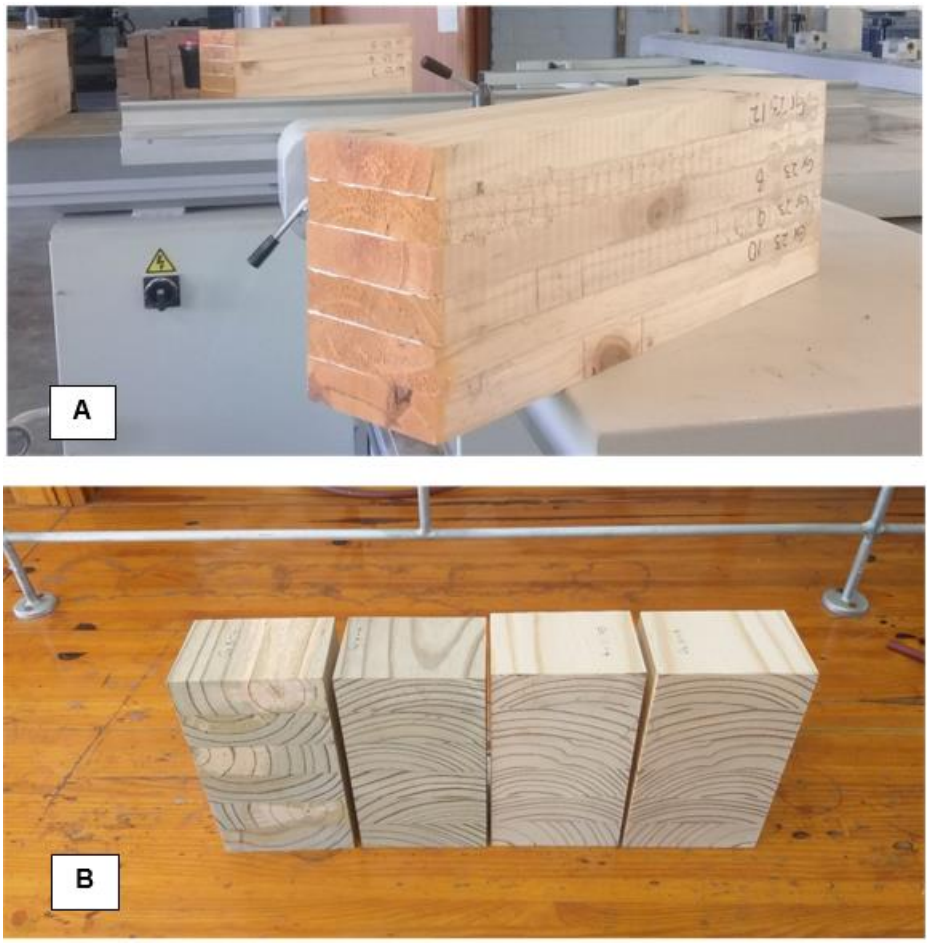


Figure 3-7: Delamination laminate (A) and 75mm test blocks (B).

After cutting the test blocks and labelling, the test block specimens were stored in a conditioning room (65% ± 5% RH, 20º ± 2ºC) for a period of 7 days before testing.

For shear test groups, each group consisted of 12 replicates and for delamination test groups of 8 replicates. Table 3-7 shows the total number of test blocks for each treatment per test.

Table 3-7: Total number of samples per treatment.

Treatment	Number of groups	Total number of shear strength test blocks	Total number of delamination test blocks
CCA	8	96	64
DOT	8	96	64
Untreated	4	48	32
TOTAL	20	240	160

3.2.8. Performance test methods

In order to fulfil the objectives of this research, the strength and durability of 1C-PUR adhesive bond lines on treated wood laminates was assessed and evaluated through standardised test methods in accordance with ASTM D905 (2008) and SANS 10183-4-2 (2009) standards.

3.2.8.1. Shear strength test

The shear strength test was conducted in accordance to ASTM D905 (2008) and performed with an INSTRON tensile testing machine using a load cell with a capacity of 50 000 N. The shearing tool had a cylindrical self-adjusting bearing, which ensured a uniform distribution of the load over the test block glueline as shown in Figure 3-8. The test machine was set to a displacement rate of 5 mm/min (cross head speed) prior to testing.

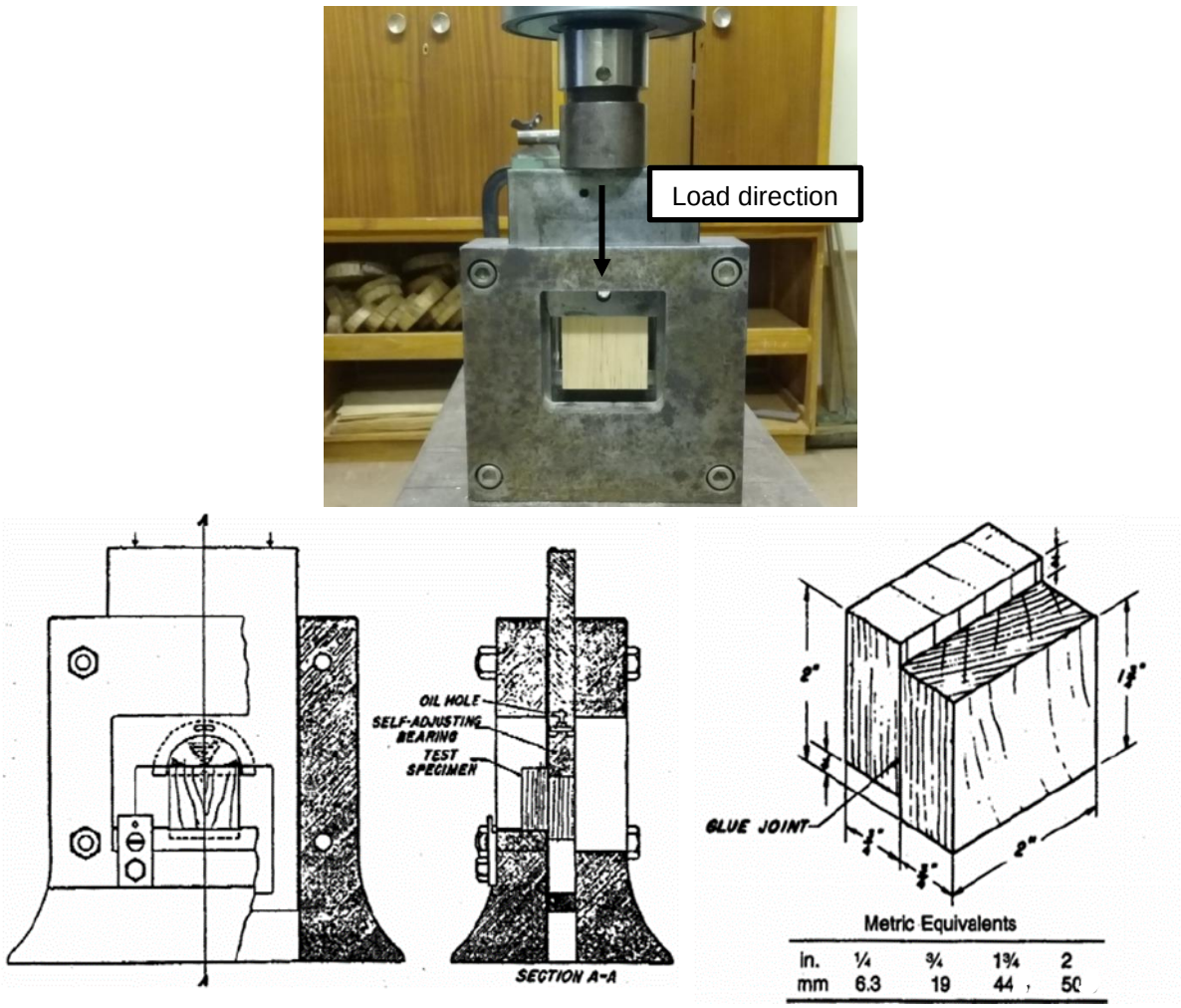


Figure 3-8: Shear testing tool.

Steps:

1. The test block was placed in the testing machine and a load with a continuous motion of a movable head of 5 mm/min was applied on the cross section of test specimen (on one adherend) until failure.
2. The maximum load applied (F_{max}) was recorded at the time of failure and the shear strength (N/mm^2) was calculated with using Equation 2:

$$fv = \frac{F_{max}}{A} \tag{2}$$

f_v = shear strength [N/mm^2]

F_{max} = maximum load applied on test sample [N]

A = cross section of the test sample glued area [mm^2] (shear area = 1885 mm^2)

After determining the maximum shear strength of the blocks, wood failure percentage (see Figure 3-9) was visually estimated and rounded off to the nearest 5%, using a transparent grid on both faces in order to understand the failure mode (on whether failure was on the wood or adhesive).

Figure 3-9: Wood failure evaluation of shear strength blocks.

3.2.8.2. Resistance to delamination

The impregnating-drying cycle delamination test was conducted in accordance to SANS 10183-4-2 (2009) standard. The delamination test consisted of three impregnating-drying cycles (high temperature procedure for Type I adhesive) and was conducted as follows:

1. 75mm thick test blocks were weighed to the nearest gram and the length of all bond lines on the end grains were measured.
2. The blocks were placed 5mm apart in a pressure cylinder in order to ensure that the end grain surfaces of the blocks are exposed. A cylindrical metal weight was placed on top of the blocks to prevent them from floating during pressure application. The test blocks were then fully submerged with water of a temperature between 10 - 25ºC.
3. The door was closed firmly and an initial vacuum of 25 kPa was applied and maintained for 15 minutes. The vacuum was then released and a pressure of 625 kPa applied for 60 minutes.
4. With the test specimens still completely immersed, the vacuum-pressure cycle was repeated once again to give a two-cycle impregnating period totalling 2h 30min (SANS 10183-4-2, 2009).
5. The test blocks were then removed from the pressure vessel and placed in a drying oven (see Figure 3-10) for 20 hours at 65 to 68ºC temperature, with a relative humidity of 10 to 15% and air velocity between 2 to 2, 50 m/s. The drying at high temperatures and low relative humidity causes a moisture gradient in the wood, which leads to high stresses in the bond line (Schmidt and Knorz, 2010).
6. After the drying period had elapsed, the test blocks were re-weighed. The test blocks had to be dried to within 100 – 110% above their initial weight for an impregnating-drying cycle to be considered complete. In some cases, the required mass of the dried test blocks was achieved in less than 20 hours, especially for sapwood test blocks. As such, the test specimens were subjected to a weight inspection after 15 hours of drying, to ensure that the test specimens are not over-dried.
7. The entire impregnating-drying cycle was repeated two more times to give a total of 3 complete cycles. The weight of each test block was recorded after every impregnating-drying cycle and the total drying time.

Figure 3-10: Delamination test blocks in the drying oven.

Measurement and calculation of delamination:

After the completion of the 3 impregnating-drying cycles, the lengths of the glueline openings were measured on both end grain surfaces of the test block. The test blocks were measured within one hour after their removal from the drying oven, as delays can close up areas of poor bond due to the specimen picking up the moisture in the air. Figure 3-11 shows delamination blocks after three impregnating-drying cycles. For accurate analysis, the openings on the gluelines were assessed under a 10x magnifying glass. For each test block, both the total delamination (*Delam*$_{tot}$) and the maximum delamination (*Delam*$_{max}$) of an individual glueline were calculated by relating delamination length to the glueline lengths (mm) (Knorz *et al*., 2017). The delamination results were expressed in percentages and the allowed proportions of delamination were considered valid if *Delam*$_{max}$ ≤ 30% *and Delam*$_{tot}$ ≤ 10% after three impregnating-drying cycles.

The total delamination (D_{tot}) of a single block, which expresses the proportion of the delamination length ($l_{tot,delam}$) of all gluelines on both end-grain surfaces to the total length of all gluelines of a block ($l_{tot,glueline}$) (Steiger *et al*., 2014), was calculated according to Equation 3:

$$D_{tot} = 100 \times \frac{\sum l_{tot,delam}}{\sum l_{tot,glue\ line}}\ [\%] \qquad (3)$$

The maximum delamination (D_{max}) in any single glueline representing the proportion of the largest delamination length on both end-grain surfaces of a single glueline ($l_{max,delam}$) to the total length on both end-grain surfaces of the same glueline ($l_{glueline}$) (Steiger, Arnold and Risi, 2014) was calculated according to Equation 4:

$$D_{max} = 100 \times \frac{\sum l_{max,delam}}{\sum l_{glue\ line}}\ [\%] \qquad (4)$$

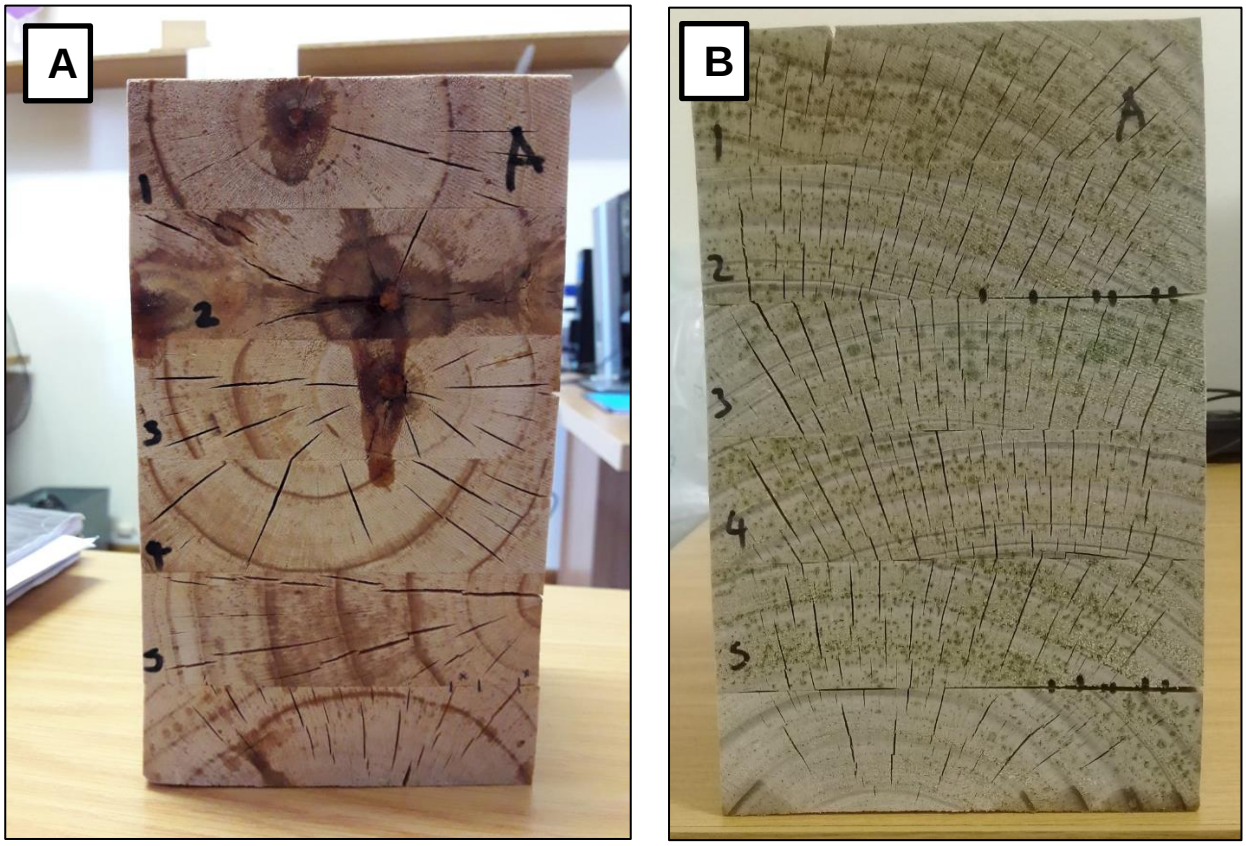


Figure 3-11: Heartwood untreated (A) and sapwood CCA-treated (B) delamination test blocks (with marked delamination openings) after three impregnating-drying cycles.

Conditions for valid glueline openings as delamination were valid when:

1. A cohesive crack in the adhesive layer,
2. A failure of the glueline precisely between the adhesive layer and the wood lamella it is adhered to, and;
3. A wood failure which is invariably within the first layers of cells beyond the adhesive layer, in which the fracture path is not influenced by the grain angle and growth-ring structure (SANS 10183-4-2, 2009).

The measurement of glueline openings excluded defects such as knots, resin pockets and grade defects and also failures in the latewood area of an annual ring which were adjacent and parallel to the glueline (SANS 10183-4-2, 2009).

3.2.9. Data analysis

R studio and Statistica 13 statistical software were used for analysis of results. A four-way ANOVA was employed for the analysis of retention results in subsection 4.2., with density, wood type, chemical and concentration as input factors.

A three-way ANOVA was carried out when analysing the effect of preservatives (CCA, DOT) and wood properties (density, wood type) on shear strength (f_v), wood failure percentage (WFP) and delamination (D_{tot}) in subsections 4.3.1 to 4.4.2. Each preservative was compared against the untreated results.

A one-way ANOVA was also carried out to compare the means of the preservatives on shear strength, wood failure percentage, and delamination.

The two primary assumptions of ANOVA - normality distribution (using the Shapiro-Wilk's test) and homogeneity of variance (using the Levene's test) were tested before proceeding with analysis. In cases where the assumptions were violated, the data was transformed using the Box Cox Transformation and Tukey's ladder of powers transformation. In some instances where the ANOVA for transformed data identified the same significant factors as the untransformed data ANOVA, the ANOVA results and interaction plots displayed were from untransformed data, since values that have been transformed provide no true meaning (Dugmore, 2018) and in most cases cannot be interpreted.

In addition, a Tukey's HSD post-hoc (*pairwise comparison*) follow up test was carried out when interactions were found to be statistically significant. The ANOVA analysis was carried out at a significance level (α) of 0.05. Graphs of the highest order of significances were displayed for the findings.

Chapter 4 : Results and Discussions

In this chapter the statistically analysed experimental results for retention rate, shear strength, wood failure and delamination are presented.

The results are outlined into two subsections as per objective:

- Evaluating the influence of wood properties (sapwood, heartwood and density) on retention rate (*see section 4.2*)
- Evaluating the effect of CCA and DOT wood preservatives and wood properties on bond performance and specifically the shear strength and delamination (*see section 4.3 and 4.4*)

4.1. Overview of shear strength, wood failure and delamination results

The shear strength, wood failure and delamination results are summarized in Table 4-1, Table 4-2 and Table 4-3, with detailed information of the groups. The results are also presented in the form of mean boxplots (in Figure 4-1 for shear strength, Figure 4-2 for wood failure and Figure 4-3 for delamination) to help visualize the distribution of the mean values and enable comparison. For the delamination results, only total delamination was reported because according to Dugmore (2018), total delamination is the most critical criterion for determining delamination. (See APPENDIX A, B for detailed information on the groups data).

Table 4-1: CCA groups mean shear, WFP and total delamination results.

Group	1	2	3	4	5	6	7	8
CCA	2%	4%	2%	4%	2%	4%	2%	4%
Wood type	SW*	SW	SW	SW	HW*	HW	HW	HW
Density	Low	Low	High	High	Low	Low	High	High
Shear strength (N/mm^2)	9.28	10.0	10.48	11.43	9.34	10.76	10.25	11.87
WFP	74%	75%	71%	93%	74%	73%	68%	48%
Delamination	1.37%	1.61%	1.61%	1.61%	1.68%	2.48%	1.20%	5.00%

*SW – Sapwood, HW - Heartwood

Table 4-2: DOT groups mean shear, WFP and total delamination results.

Group	9	10	11	12	13	14	15	16
DOT	1.67%	3.3%	1.67%	3.3%	1.67%	3.3%	1.67%	3.3%
Wood type	SW	SW	SW	SW	HW	HW	HW	HW
Density	Low	Low	High	High	Low	Low	High	High
Shear strength (N/mm^2)	10.09	9.03	10.07	10.13	9.71	11.45	11.04	10.29
WFP	88%	88%	91%	76%	93%	90%	92%	89%
Delamination	0.99%	2.19%	1.09%	1.35%	1.66%	1.27%	1.34%	2.30%

Table 4-3: Control (untreated) groups mean shear, WFP and total delamination results.

Group (control)	17	18	19	20
Wood type	SW	SW	HW	HW
Density	Low	High	Low	High
Shear strength (N/mm^2)	9.83	10.19	10.24	11.20
WFP	89%	88%	69%	62%
Delamination	1.65%	1.97%	3.22%	1.73%

Shear strength test results: the shear strength mean values for all groups were above 9.0 N/mm^2 (see Table 4-1, Table 4-2, Table 4-3 and Figure 4-1). This indicated good bond performance despite the presence of preservatives, as the gluelines met the minimum requirements of 6 N/mm^2 for EN 14080 (refer to Table 4-7). Interestingly, group 8, which had heartwood blocks of high density and treated with an aqueous solution of 4% CCA, exhibited the highest shear strength mean of 11.87N/mm^2. These results may suggest that CCA and DOT preservatives and wood properties (density and sapwood/heartwood) do not affect bond strength to the extent of disqualifying them from structural use as it has been reported in literature.

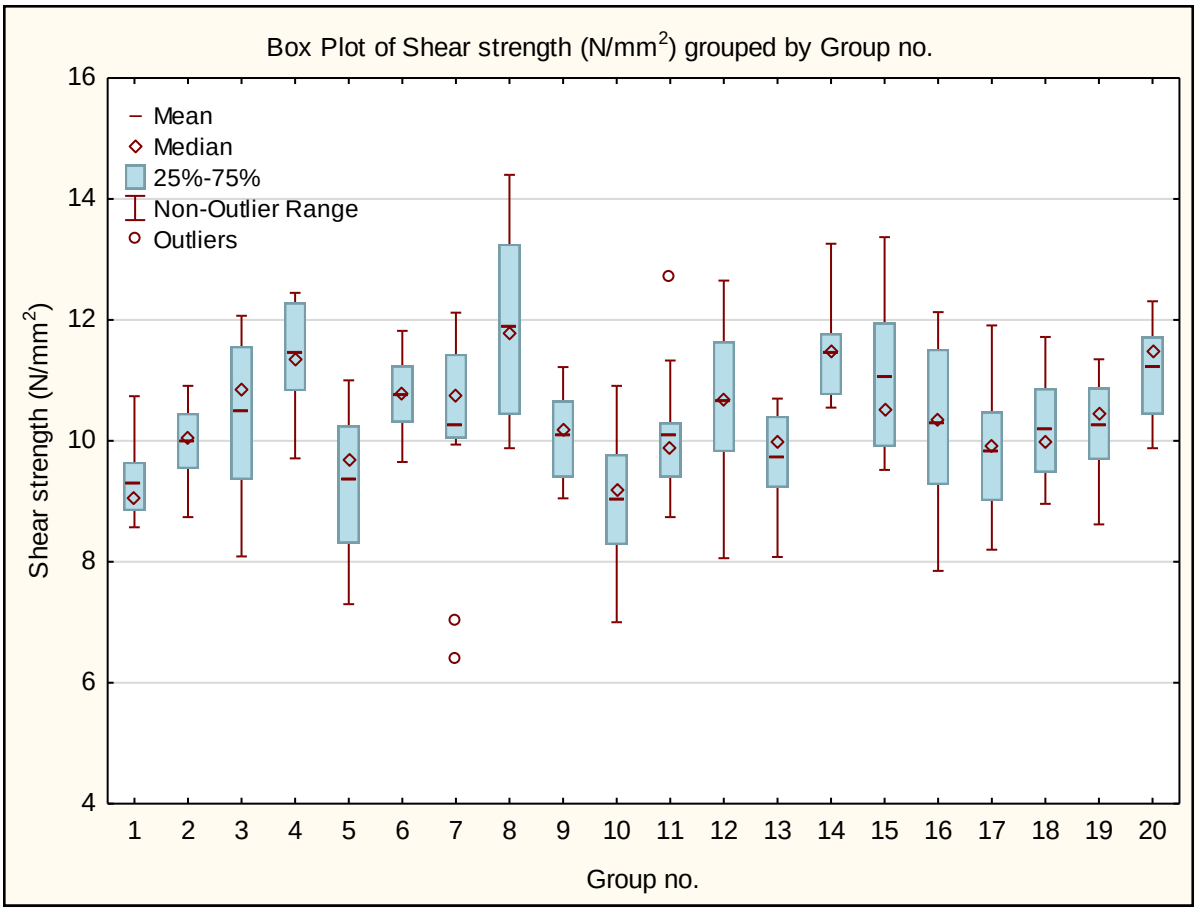


Figure 4-1: Boxplot of mean shear strength values grouped by group number.

Wood failure test results: Based on the wood failure data presented in Table 4-1, Table 4-2, Table 4-3 and visual distribution of the wood failure bars in Figure 4-2, overall the DOT-treated test blocks displayed a higher wood failure percentage in comparison to CCA-treated and control (untreated) test blocks. The high wood failure observed for DOT-treated blocks may serve as confirmation that DOT preservative had minimal effect on the PUR glueline. While Group 8 displayed the highest shear strength in Figure 4-1, in terms of wood failure, it displayed the lowest wood failure of 48%.

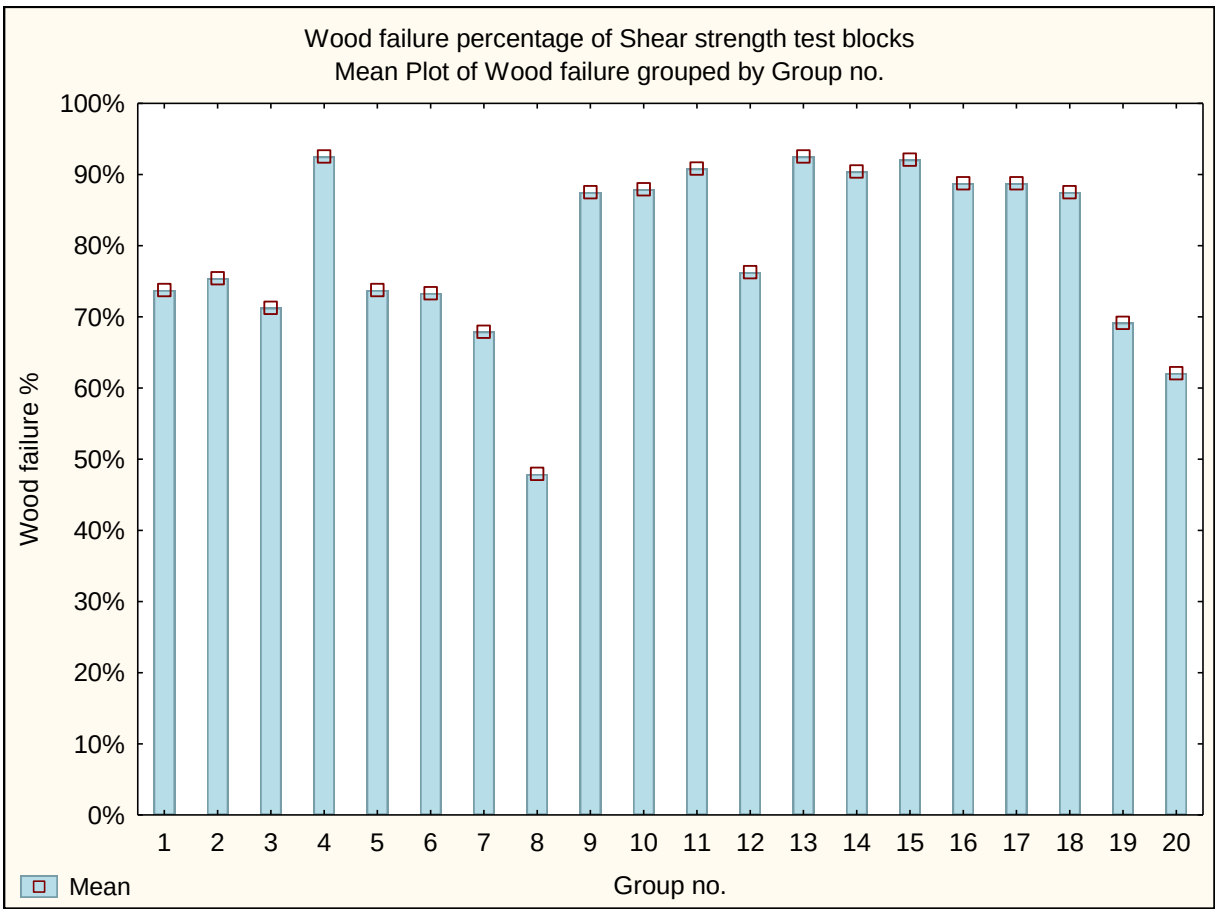


Figure 4-2: Mean plot of wood failure percentage values grouped by group number.

Delamination test results: the delamination results in Figure 4-3 demonstrate that CCA and DOT-treated and control (untreated) test blocks met the requirements of EN 14080 (2013), as they did not exceed the 10% total delamination limit in length. As the data in Figure 4-3 and Table 4-2 shows, group 9 (1.67% DOT x sapwood x low density) had the lowest total delamination value of 0.99%. Interestingly, group 8, which had the highest shear strength of 11.87 N/mm^2 (see Table 4-1), displayed the highest total delamination of 5% (see Figure 4-3 and Table 4-1). These results seem to suggest that there was a poor relationship between shear strength and delamination in terms of bond performance (*high shear strength does not necessarily mean bond lines will be highly resistant to delamination*). Furthermore, some of the heartwood groups (group 6, 8,16, 19) displayed

high total delamination values, indicating that heartwood extractives may have caused some interference with bonding.

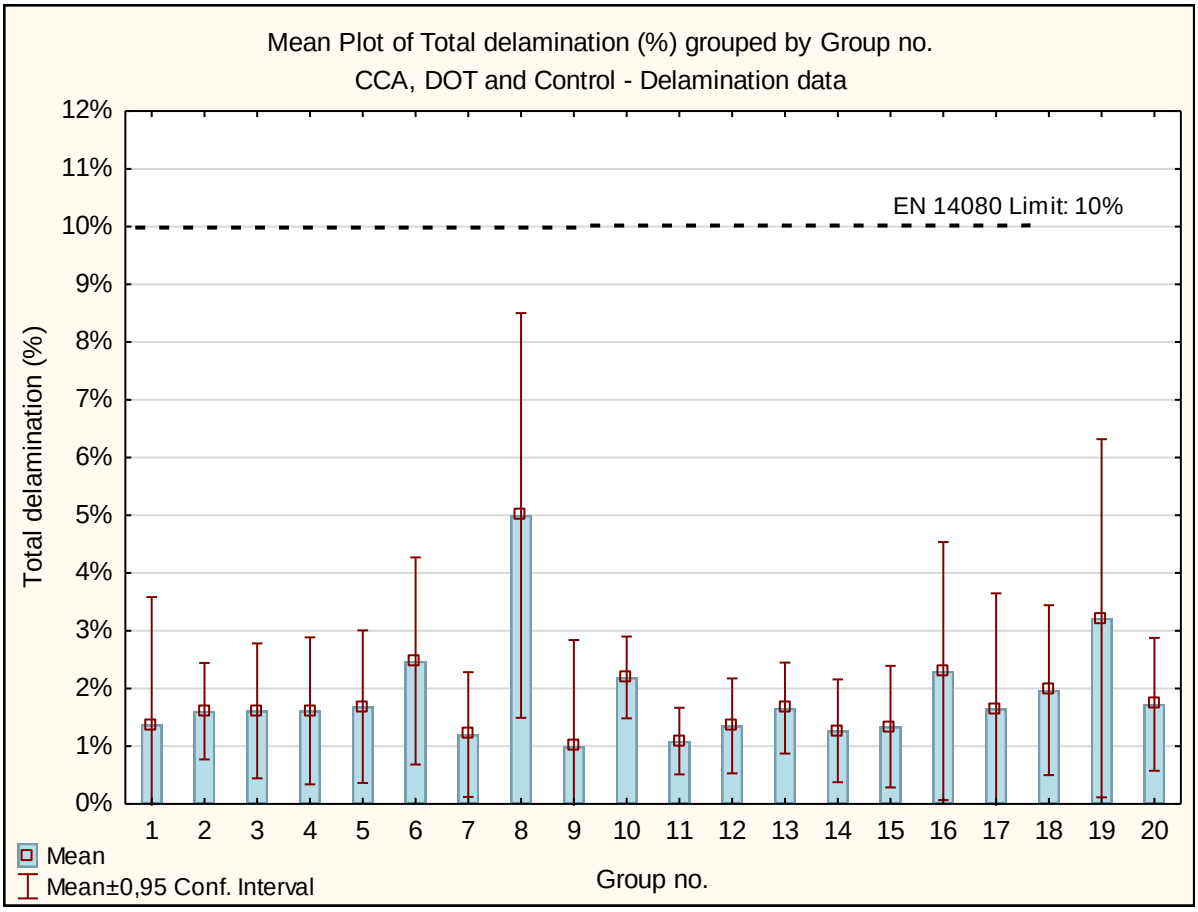


Figure 4-3: Mean plot of delamination values grouped by group number. The error bars indicate the variability/spread of the delamination data.

4.2. Relationship between wood properties and retention rate

The results related to the relationship between the wood properties (density and sapwood/heartwood ratio) and retention rate are presented and discussed in this section.

Table 4-4: Retention rate of CCA and DOT preservatives for shear and delamination samples

Shear and delamination samples							
Group	**Chemical**	**Concentration**	**Wood type**	**Density**	**Targeted retention**	**Shear samples mean retention rate (kg/m³)***	**Delamination samples mean retention rate (kg/m³)****
1	CCA	2%	Sapwood 100%	Low density	6 kg/m³	7,71	7,99
3	CCA	2%	Sapwood 100%	High density		6,97	8,01
5	CCA	2%	Heartwood>35%	Low density		8,17	7,65
7	CCA	2%	Heartwood>35%	High density		7,12	5,55***
2	CCA	4%	Sapwood 100%	Low density	12 kg/m³	15,79	14,78
4	CCA	4%	Sapwood 100%	High density		15,80	16,49
6	CCA	4%	Heartwood>35%	Low density		14,59	16,30
8	CCA	4%	Heartwood>35%	High density		14,46	13,03
9	DOT	1,67%	Sapwood 100%	Low density	5 kg/m³	7,22	7,67
11	DOT	1,67%	Sapwood 100%	High density		6,80	6,66
13	DOT	1,67%	Heartwood>35%	Low density		5,64	5,48
15	DOT	1,67%	Heartwood>35%	High density		5,04	5,25
10	DOT	3,3%	Sapwood 100%	Low density	10 kg/m³	14,15	14,79
12	DOT	3,3%	Sapwood 100%	High density		12,48	13,08
14	DOT	3,3%	Heartwood>35%	Low density		11,57	14,94
16	DOT	3,3%	Heartwood>35%	High density		9,16***	10,12

**Sample dimensions: 20mm x 51mm x 310mm ; ** Sample dimensions: 32mm x 110mm x 500mm; ***Target retention not met*

Statistical Analysis

The majority of the group's samples in the experiment met and exceeded the targeted retention rates except for group 16 shear samples and group 7 delamination samples (see Table 4-4).

A four-way factorial ANOVA was carried out in order to detect if the retention rate was influenced by the wood factors (density and wood type) and chemical preservatives. A Tukey's HSD post-hoc (pairwise comparison) follow-up test was also carried out when interactions were found to be statistically significant. The Tukey's HSD post-hoc test assisted in identifying which specific group's mean combinations were significantly different. Highest order interactions were interpreted in cases where interactions were significant.

In order to compare retention results of specimens treated at different concentration rates of the chemicals, the retention rate results for the high concentration levels (4% CCA and 3.3% DOT), were halved before carrying out the ANOVA. The assumption was that retention rates of the high concentration groups will simply be double that of the low concentration group ONLY, because the concentration levels was double that of low concentration groups. In this way the effect of density, wood type and chemical could be analysed on all the specimens together.

See ANOVA table below:

Table 4-5: ANOVA table for retention rate results

Effect	Univariate Tests of Significance, Effect Sizes, and Powers for Retention (Retention data (Shear and Delamination)) Sigma-restricted parameterization Effective hypothesis decomposition							
	SS	Degr. of freedom	MS	F	p	Partial eta-squared	Non-centrality	Observed power (alpha=0.05)
Intercept	13671.83	1	13671.83	8804.742	0.000000	0.970033	8804.742	1.000000
{1}Chemical	97.22	1	97.22	62.610	0.000000	0.187114	62.610	1.000000
{2}Wood type	74.98	1	74.98	48.288	0.000000	0.150764	48.288	1.000000
{3}Density	48.24	1	48.24	31.067	0.000000	0.102510	31.067	0.999837
{4}Concentration	2.37	1	2.37	1.524	0.218014	0.005573	1.524	0.233517
Chemical*Wood type	8.21	1	8.21	5.288	0.022228	0.019071	5.288	0.629883
Chemical*Density	2.50	1	2.50	1.607	0.205959	0.005874	1.607	0.243644
Wood type*Density	17.07	1	17.07	10.992	0.001040	0.038840	10.992	0.910470
Chemical*Concentration	0.07	1	0.07	0.043	0.836440	0.000157	0.043	0.054871
Wood type*Concentration	5.40	1	5.40	3.480	0.063187	0.012633	3.480	0.459823
Density*Concentration	0.07	1	0.07	0.045	0.831278	0.000167	0.045	0.055190
Chemical*Wood type*Density	7.08	1	7.08	4.558	0.033651	0.016483	4.558	0.566548
Chemical*Wood type*Concentration	1.11	1	1.11	0.716	0.398309	0.002624	0.716	0.134535
Chemical*Density*Concentration	11.13	1	11.13	7.166	0.007881	0.025670	7.166	0.760384
Wood type*Density*Concentration	3.61	1	3.61	2.323	0.128649	0.008468	2.323	0.329765
1*2*3*4	2.40	1	2.40	1.546	0.214866	0.005650	1.546	0.236097
Error	422.36	272	1.55					

The two highly significant interactions (at $p<0.001$) are discussed below. The ANOVA shows that concentration was not significant as a main factor, which partially support the decision to analyse all concentration level specimens together .

Wood type x Density interaction:

The wood type and density interaction was statistically significant at a 5% significance level (see ANOVA Table 4-5).

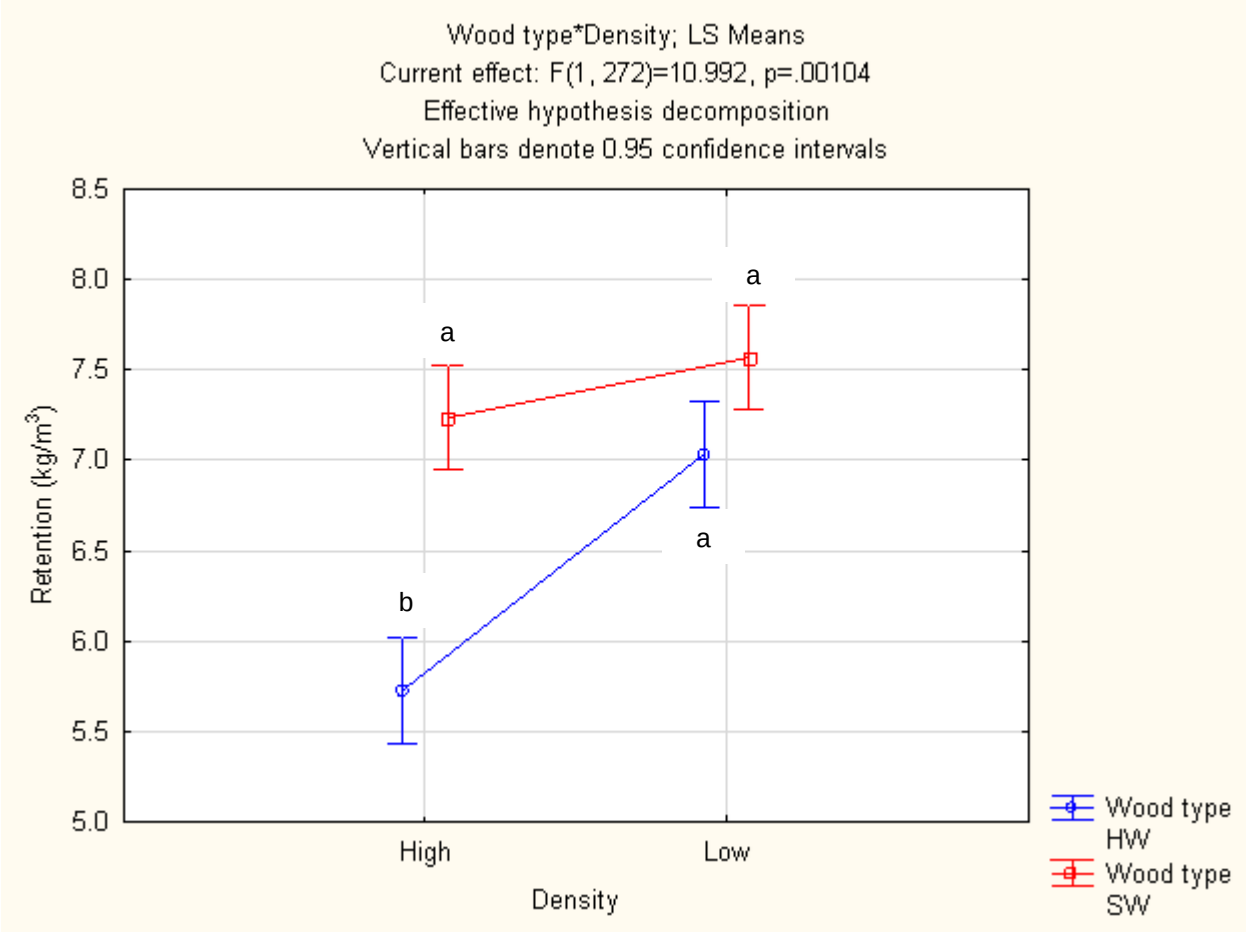


Figure 4-4: The interaction between wood type and density for retention rate (kg/m^3) of CCA and DOT preservative treated specimens.

The interaction plot in Figure 4-4 shows that the mean retention rate in high density samples was lower than that in low density samples in both heartwood and sapwood. Moreover, retention in heartwood samples appeared to be more affected by density, since high-density heartwood samples were the only ones significantly different to the other groups. One possible explanation for this phenomenon is that wood with high density has reduced void volume or lumen size (Guo *et al.*, 2002; Halverson and Lebow, 2011); moreover, when thick cell walls (high density) are present in heartwood which is known to have high extractive content deposited in the pit membranes (Wang and De Groot, 1996; Wen, Kang and Park, 2014; Tarmian *et al.*, 2020), the combination of these anatomical properties or features are bound to reduce the flow of preservative solution and limit the deposition of preservative salts in the cell lumina. Metsä-Kortelainen *et al*. (2006) and Wang and De Groot (1996) explained that certain anatomical features in heartwood such as the increased rate of irreversible aspirated pits on the tracheid cell walls, encrustation of the pit membranes, reduced pore sizes and the amount and type of extractives deposited on pit membranes during the formation of heartwood often reduce the permeability in heartwood. Tripathi (2012) also explained that the presence of heartwood can affect the physical properties and permeability of wood as the extractives contained in heartwood can infiltrate completely into the cell walls or may occur as surface deposits or plugs in cell lumina. In addition, the

physiological inactivity in the heartwood and high deposition of resinous and phenolic extractives with biotic resistance, limits or obstructs preservative flow (Wen et al., 2014). On the other hand, sapwood contains living and active tracheids and parenchyma cells which allow flow of preservative solution during treatment.

The results displayed in this experiment are similar to those of Guo *et al.* (2002), where southern pine samples of sapwood (with density of 486 kg/m^3) and heartwood (density of 476 kg/m^3) achieved retention rates of 11.2 and 7.7 kg/m^3, respectively, when treated with a 2% CCA-C solution. The authors attributed the low retention in heartwood to be caused by the presence of extractives and bordered pits in heartwood.

Furthermore, upon visual inspection of the cross section of CCA treated heartwood samples, it was visibly clear that the CCA preservative failed to penetrate the heartwood area completely (see Figure 4-5) in some heartwood samples. These observations further confirmed what was highlighted by Tarmian *et al.*, (2020) that the distribtion of preservatives such as CCA is not consistent in all parts of the wood especially in heartwood due to more pit aspiraction. Nonetheless, the failure of the preservative to penetrate the heartwood area can possibly be compensated for by the presence of high amounts of extractives which also provide natural durability or biotic resistance.

Figure 4-5: Failure of complete preservative penetration in the heartwood samples.

Chemical x Density x Concentration interaction:

The 3-way interaction (chemical x density x concentration) was statistically significant at a 5% significance level (see ANOVA Table 4-5).

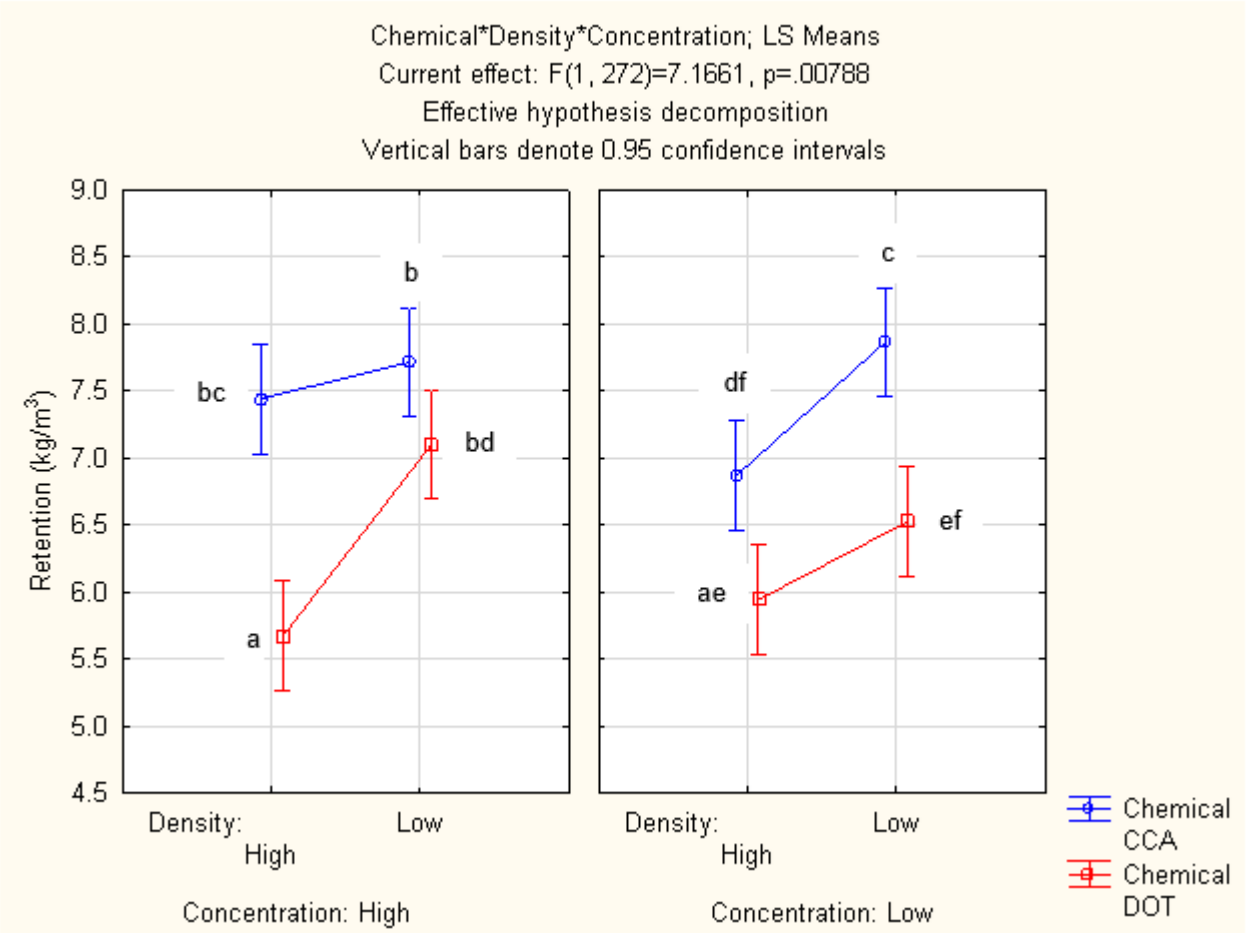


Figure 4-6: The interaction between chemical, density and concentration for retention rate (kg/m^3) of CCA and DOT preservative treated samples.

As Figure 4-6 shows, both chemicals (CCA and DOT) and their concentration levels were affected by the density. Low density samples retained more of the preservative than high density samples. These results are similar to those of Guo *et al.*, (2002), where retention was reported to decrease as density increased. Guo *et al.*, (2002) attributed the cause of low retention in denser wood to the smaller void volume/space, as it often dictates solution uptake, retention, and penetration. Similarly, Tripathi (2012) found a negative and significant correlation ($P<0.01$, $r=-0.381$) between density of meranti specimens and retention of CCA and borate-based preservatives. The low retention in high density specimens was also attributed to the void volume which decreases with increasing density. The interaction shows that DOT treated specimens are more influenced by density at high concentration levels than CCA.

Summary

Overall, the results showed that sapwood had a higher/better retention capacity than heartwood. These results further confirmed the findings of several authors that the presence of heartwood has a significant effect on

solution uptake and retention. Furthermore, having different proportions of heartwood to sapwood introduced variability in retention and solution uptake. According Lebow, Hatfield and Abbott (2005), the variability in uptake of different wood types and parts (different heartwood percentage) makes it difficult for commercial treaters to optimize treating parameters, such as treatment time and chemical consumption. Alternatively, wood that has a high proportion of heartwood can be incised to improve penetration of preservative.

Similarly, density was also found to affect the retention rate. In most cases, a negative relationship existed between density and retention. This indicates that the retention is much lower in high-density wood samples compared to low density-wood samples. Wen et al (2014) and Schultz et al (2004) also reported a similar relationship. Halverson and Lebow (2011) further highlighted that in larger specimens, the relationship between density and solution uptake becomes more nuanced as permeability becomes a greater factor. However, in this case we also tested whether the effect of specimen dimension had an influence on retention, since the shear and lamination specimens were of different sizes and found that there was no significant effect (results not shown).

Overall, these findings illustrate that it is important to understand or know the treatability behaviour/response of various parts of wood (e.g. sapwood, heartwood), anatomical characteristics (e.g. thick or thin cells walls) and size, in order to ensure the required or targeted retention and penetration is achieved during treatment.

4.2. Shear strength and WFP: effect of CCA and DOT preservatives

The shear strength test was conducted in accordance to ASTM D905 (2008), but the test results were benchmarked and based on the requirements given in EN 14080 (2013). EN 14080 (2013) defines minimum values for shear strength f_v and WFP, both in terms of individual values for a single glueline and the average values for a beam (Knorz *et al.*, 2014), as shown in Table 4-6.

Table 4-6: Minimum required values for wood failure percentage related to shear strength according to EN 14080 (2013).

Parameter	**Average values**			**Individual values**		
Shear strength f_v (N/mm^2)	6	8	$f_v \geq 11$	$4 \leq f_v < 6$	6	$f_v \geq 10$
Minimum wood failure percentage (%)	90	72	45	100	74	20

The results were given a “pass or fail” evaluation based on the corresponding value for WFP that depends on the shear strength, as given in EN 14080 (2013). It has been reported that by having both results present to be more conclusive on adhesion than based on shear strength alone (How *et al.*, 2017). Based on the overall evaluation of the test results, all groups in the experiment met the average requirements of EN 14080 (2013), both for shear strength and wood failure percentage shown in Figure 4-7. The majority of CCA treated groups displayed slightly lower values of wood failure percentage when compared to DOT treated and control groups. This might indicate that the presence of CCA metallic deposits may have affected the wood failure percentage (Cameron and Pizzi, 1985).

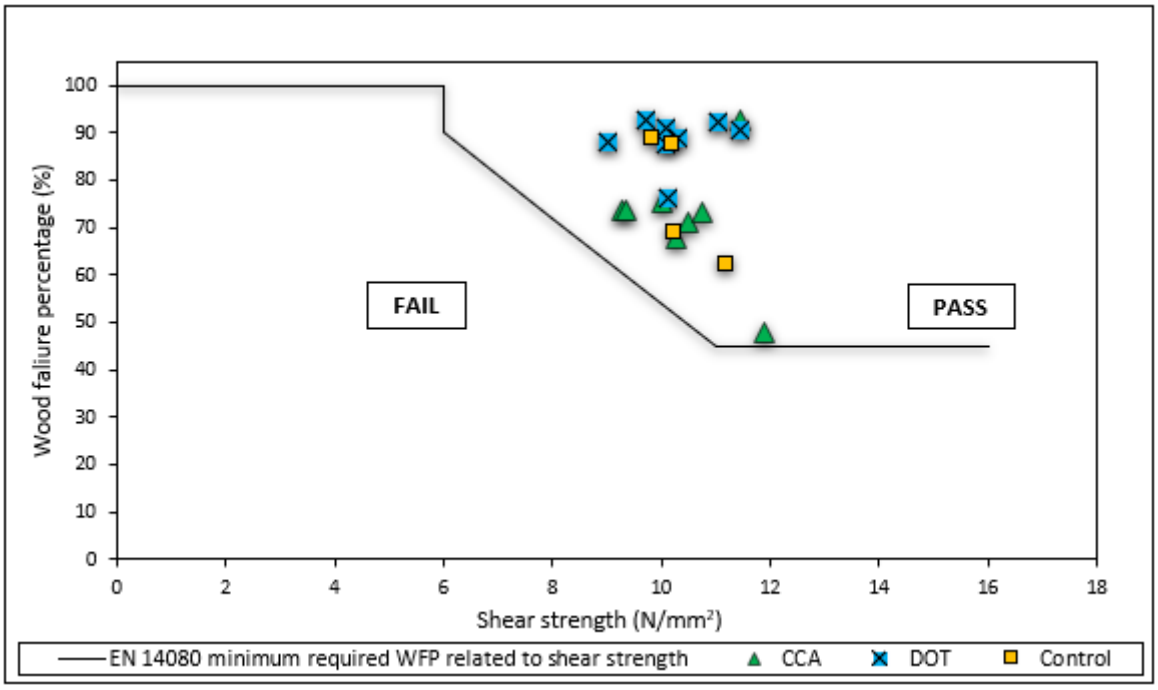


Figure 4-7: Mean block shear strength and wood failure percentage values for CCA, DOT and control groups in accordance to EN 14080 (2013) average values requirements.

4.3.1. CCA and control shear strength

A 3-way factorial ANOVA (3 x 2 x 2) was conducted in order to assess whether the presence of CCA preservative and certain wood properties (density and sapwood/heartwood) had any statistical significance on the bond strength of PUR adhesive in pine laminates in comparison to untreated samples.

The shear strength data violated the assumption of normal distribution and was transformed using the Box-Cox transformation. However, the ANOVA for transformed data identified the same significant factors as the untransformed data ANOVA. As such, the ANOVA results and interaction plots displayed will be from untransformed data. The ANOVA results are presented in Table 4-7.

Table 4-7: ANOVA shear strength results for CCA-treated and control (untreated) test blocks.

Effect	SS	Degr. of Freedom	MS	F	p
Intercept	15595,85	1	15595,85	13118,54	0,000000
Wood type	6,06	1	6,06	5,10	0,025606
Density	35,66	1	35,66	30,00	0,000000
Concentration	33,42	2	16,71	14,06	0,000003
Wood type*Density	0,00	1	0,00	0,00	0,995131
Wood type*Concentration	4,44	2	2,22	1,87	0,158803
Density*Concentration	2,27	2	1,13	0,95	0,387663
Wood type*Density*Concentration	1,64	2	0,82	0,69	0,504538
Error	156,93	132	1,19		

The main factors (wood type, density, concentration) were all statistically significant at a 5% significance level (see Table 4-7).

Concentration:

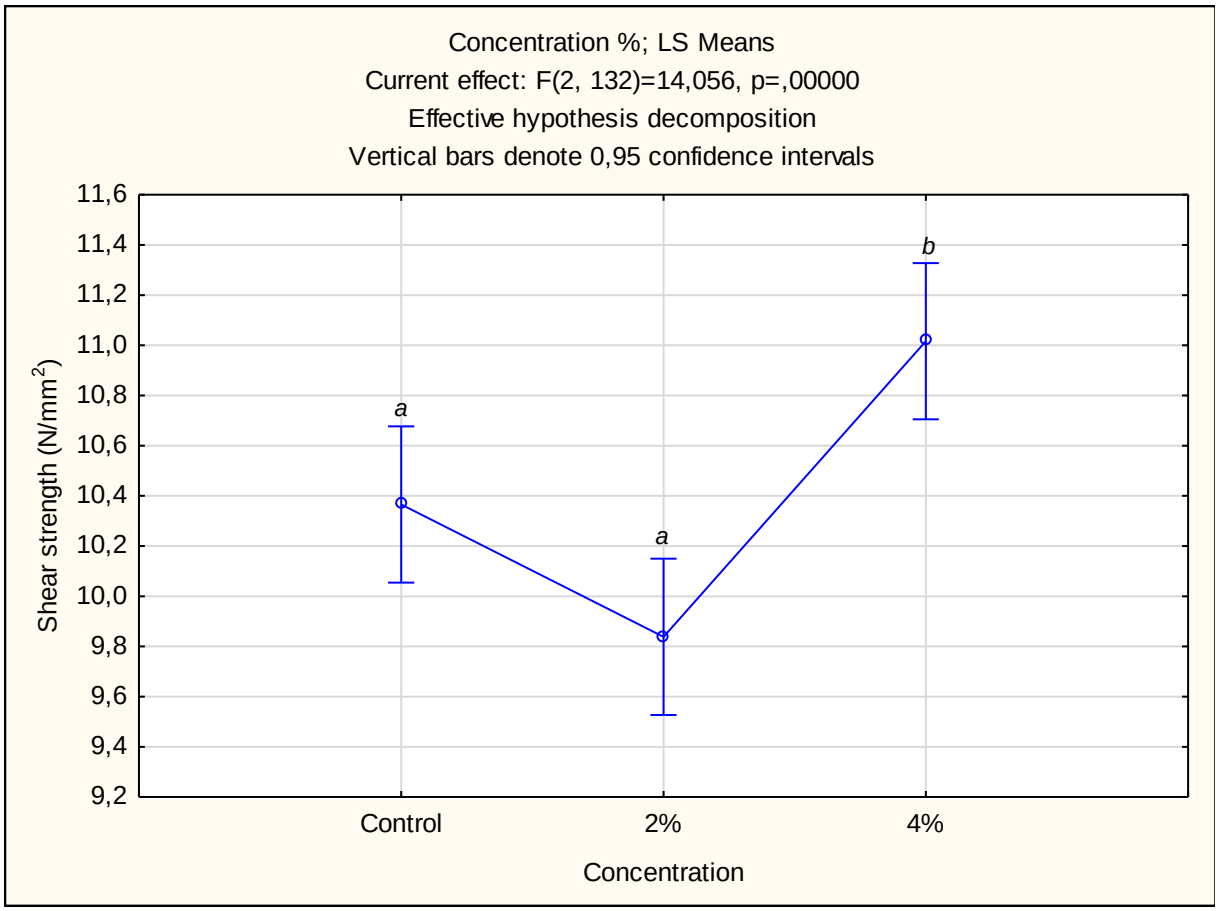


Figure 4-8: The effect of different CCA concentration levels in comparison to untreated on shear strength.

The CCA concentration levels were found to be statistically significant ($p<0.01$) at a 5% significance level (see Table 4-7).

From Figure 4-8, the control (untreated) samples displayed a slightly higher shear strength when compared to samples treated with 2% CCA, although the difference was not statistically significant. As reported in the literature, there are several studies, which found that CCA preservatives reduce adhesion strength. For instance Ozdemir *et al*. (2015) highlighted that low bond strength should be expected on wood specimens treated with 2% CCA, as CCA preservative causes poor wetting properties and increased contact angle. Similarly, Zhang *et al.* (1997) reported approximately 20% shear strength loss in CCA treated southern pine bonded with RF adhesive. Maldas and Kamdem (1998) explained that the deposition of As, Cu and Cr oxides causes the wood surface to become rougher, less polar, hydrophobic and acidic, due to the ion exchange and adsorption reactions that occur between the metals and wood. Furthermore, once the CCA metal oxides are deposited into the cell lumen during pressure treatment, there is a lack of adsorption of the adhesive onto the already heavily coated wood fibres, which may lead to reduced opportunities for stable chemical bonding between wood adherents and adhesive .

However, when the control samples were compared to the 4% CCA treated specimens (see Figure 4-8), a somewhat unexpected relationship was displayed. The 4% CCA treated specimens displayed a superior shear strength to the control, which contradicts the findings of Ozdemir *et al.* (2015), Özçifçi (2006) and several

other authors (Zhang *et al.*, 1997; Lee *et al.*, 2006). However, Cameron and Pizzi (1985) reported similar results that shear strength is unaffected by the increasing levels of CCA retention, although wood failure is reduced. The authors achieved the highest shear strength results with 32 kg/m^3 retention (out of all the retention levels tested 0, 16, 20, 32 kg/m^3) at a spread rate of 200 g/m^2 of PRF. Cameron and Pizzi (1985) explained that the fixed chromium in wood forms a strong, stable and irreversible complex with the adhesive, resulting in higher surface wetting and giving a very high strength.

Tascioglu *et al*. (2003) also reported similar results when evaluating the adhesive shear strength of CCA pre-treated southern yellow pine and FRP composite bonded interface. Their results showed that CCA concentration levels from 2.5%, 5% and 10% resulted in higher shear strength when compared to the control, whilst a 1% CCA concentration level resulted in lower shear strength in comparison to control. These results displayed a similar pattern as those reported for this study (see Figure 4-8).

Another explanation for these conflicting results could be explained by the findings of Tascioglu *et al.* (2004). The authors conducted a surface energy analysis using a surface electron microscope (SEM) and an energy dispersive X-ray analysis (EDX) on southern pine samples treated with CCA at 1%, 2.5%, 5% and 10%. Interestingly, the total surface energy of southern yellow pine was found to increase with increasing concentration levels of CCA. The increased surface energy was attributed to the chemical modification of the wood surface by the high surface energy metallic salts. An accumulation of these high surface energy metallic salts utilizing a SEM microscope were observed (Tascioglu *et al.*, 2004). Based on the surface energy analysis results of Tascioglu *et al.* (2004), this may have caused the increased shear strength in specimens treated with 4% CCA in this experiment, since most adhesives are water based and need wood with high surface energy to be able to wet and penetrate wood (Frihart, 2004). Kamke and Lee (2007) also highlighted that greater surface energy of the wood promotes greater wetting and penetration of the adhesive. However, since no surface energy analysis was conducted in this experimental study, these assumptions cannot be confirmed, and thus, further research will be required in future.

Another possible explanation for these contrary results could be that the effect of the preservative deposits on adhesion bond formation might have been eliminated through surface planing prior to adhesive application. According to Maldas & Kamdem (1998), Özçifçi (2006) and Tascioglu et al. (2003), CCA deposits increase surface roughness and cause higher contact angles and as such, the intimate contact between the wood adherents and the adhesive molecules is obstructed. But since the lamellas were surface-planed before adhesive application, this might have improved wettability thorough the removal of preservative deposits on the surface and lowered the contact angle. Hse and Kuo (1988) reported that freshly planned wood surfaces have highly polar surfaces to which adhesives bond most efficiently.

Wood type:

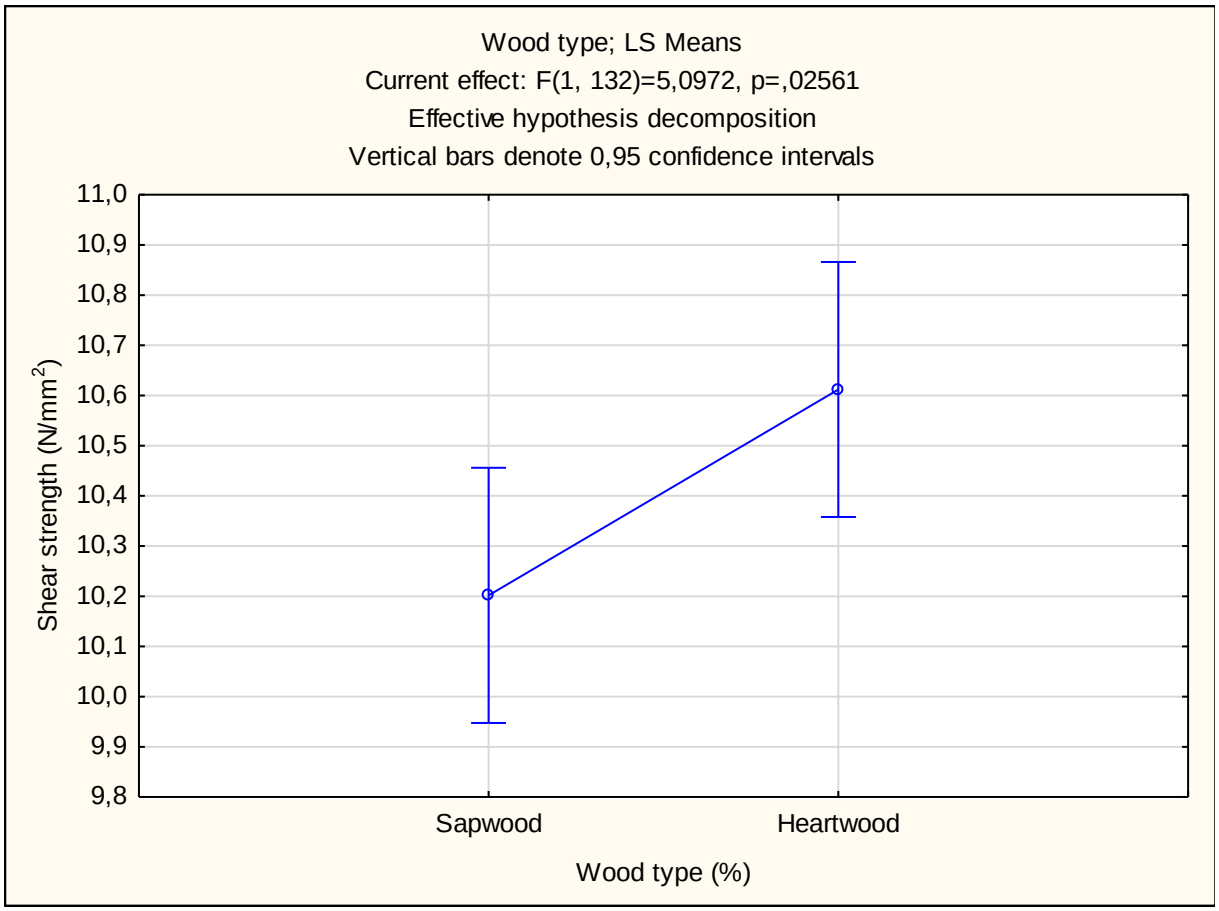


Figure 4-9: Effect of wood type in CCA treated and untreated samples on shear strength.

The results indicated that the shear strength difference between heartwood and sapwood was statistically significant ($p<0.05$) (see Table 4-7).

Figure 4-9 displays heartwood having a higher shear strength in comparison to sapwood. Based on research of untreated wood, Clauß et al. (2011), Kaygin & Tankut (2008) and Roffael (2016) found that the high level of extractives in heartwood interfere with adhesion and reduces the bond strength, due to the extractives blocking the reaction sites and preventing the anchoring of the adhesives. Hse and Kuo (1988) further highlighted that during the drying process of wood, the heartwood extractives migrate to the wood surface, which alters the properties of wood as an adherent. However, in this study the results were contrary to what has been reported.

One possible explanation for higher shear strength of treated heartwood samples in our study could be that over-penetration of the adhesive might have occurred in the sapwood since the lumens are not filled with extractive deposits and have no aspirated bordered pits. As a result, this might have led to the bond line not having enough adhesive to form strong mechanical interlocking between the sapwood substrates.

Another possible explanation for the higher shear strength in samples classified as heartwood, could be that the different proportions/ratio's of sapwood/heartwood (35 – 100%) in heartwood samples influenced the results. To explain this, the glueline might have been along the sapwood proportion, as illustrated in Figure 4-10. Which meant that as much as the samples were classified as heartwood, but the adhesive might have

been applied and penetrated on the sapwood proportion and thus, minimizing the effect of heartwood tissues on adhesion.

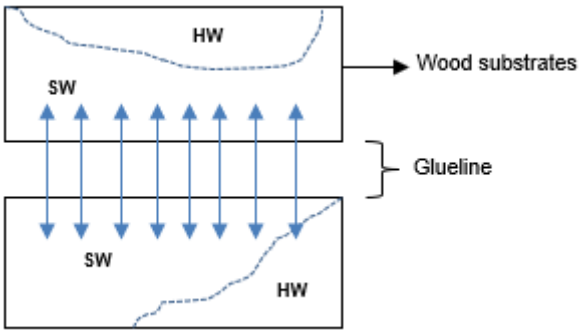


Figure 4-10: Penetration of adhesive in samples classified as heartwood.

Another possible explanation for higher shear strength of treated heartwood samples in our study could be that the effect of extractives might have been eliminated through surface planing prior to adhesive application. According to Hse and Kuo (1988), a light planing is the most effective method of removing the deleterious effect of extractive contamination. Planing not only removes extractives contaminating the wood surface, it also exposes a fresh and highly polar surface, to which adhesives bond most efficiently (Hse and Kuo, 1988).

Roffael (2016) also explained that certain extractives in heartwood are of high acidity and tend to accelerate or decelerate the curing of adhesives depending on the pH of the adhesive. Roffael (2016) further added that some extractives are highly reactive towards the main components of the adhesive and can change the bond formation. This highlights that certain extractives may have influenced bond formation and led to the high shear strength for heartwood specimens. However, extractive characterisation (composition, quantity and pH of extractives etc.), which was outside the scope of this experiment, would be required to confirm these assumptions.

Density:

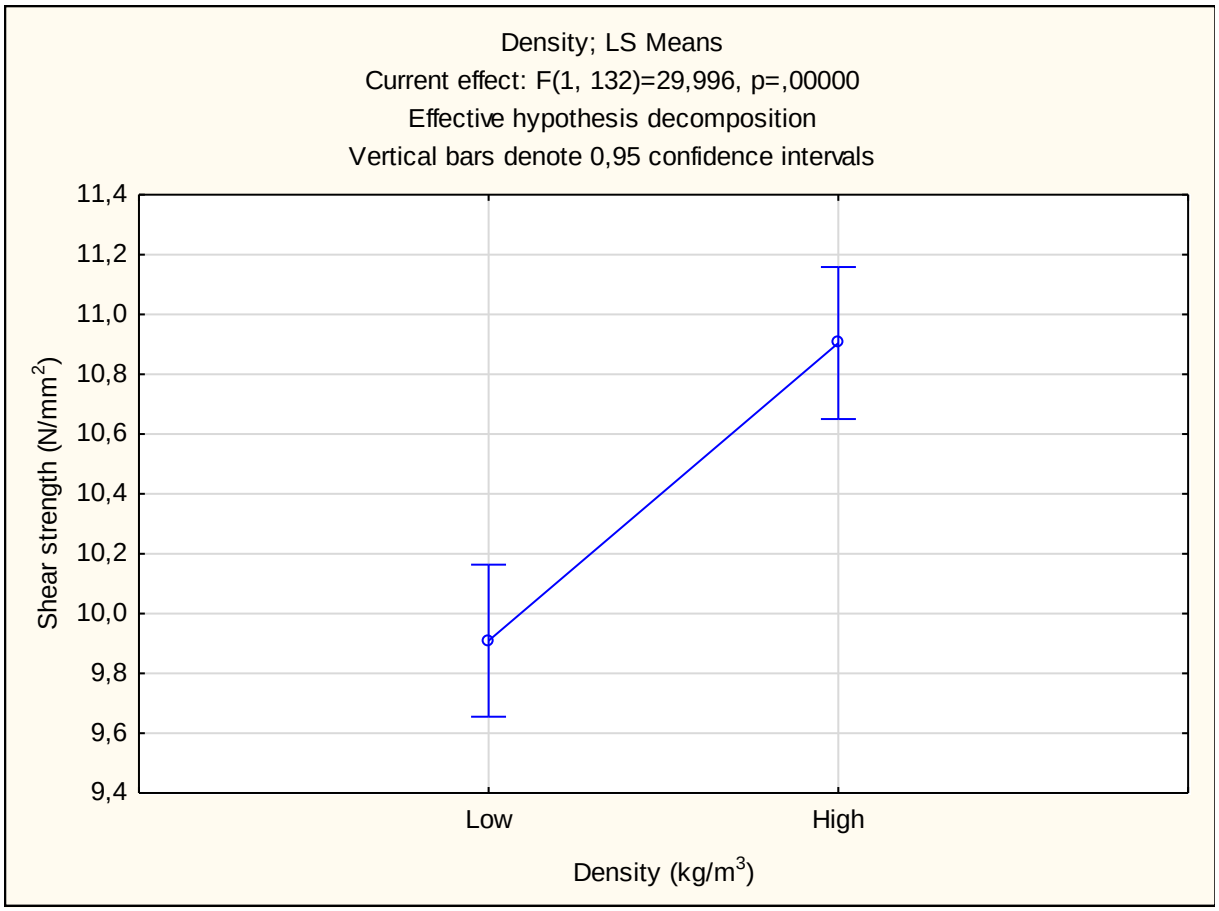


Figure 4-11: Effect of wood density on shear strength of CCA treated and control (untreated) samples.

The results indicated that the shear strength difference between low and high density was statistically significant at a 5% significance level (see Table 4-7).

Figure 4-11 shows high density having a greater shear strength in comparison to low density wood. These results displayed the expected outcomes and confirmed the findings of several authors that denser woods often have higher values of shear strength. Vick (1999) reported that the strength of adhesive bonds increases with wood density. This is because of the thick-walled cells in high density woods which are capable of withstanding much greater stress than thin-walled cells of low density (Vick, 1999; Frihart and Hunt, 2010).

Hunt *et al.*, (2019) also explained that since low density woods usually have more voids and large lumina openings, this allows better adhesive flow into the wood, however, this can lead to over-penetration, where too much adhesive flows away from the bond line. This results into a starved bond line and can consequently lead to reduced bond strength due to the thin glueline failing to form strong surface adhesion and mechanical interlocking.

Wood failure percentage

A 3-way factorial ANOVA (3 x 2 x 2) was carried out to test the influence of CCA preservative on wood failure percentage in comparison to untreated samples. The wood failure percentage data violated the assumption of normal distribution and was transformed using the Box-Cox transformation. However, the transformed data ANOVA in Table 4-8 identified the same significant factors and interactions as the untransformed data ANOVA.

As such, the ANOVA results and interaction plots displayed will be from untransformed data. The ANOVA results are presented in Table 4-8.

Table 4-8: ANOVA wood failure percentage results for CCA treated and control blocks.

Effect	SS	Degr. of Freedom	MS	F	p
Intercept	780277,8	1	780277,8	1740,544	0,000000
Concentration	776,4	2	388,2	0,866	0,423034
Wood type	9025,0	1	9025,0	20,132	0,000016
Density	625,0	1	625,0	1,394	0,239825
Concentration*Wood type	3616,7	2	1808,3	4,034	0,019934
Concentration*Density	0,0	2	0,0	0,000	1,000000
Wood type*Density	2669,4	1	2669,4	5,955	0,016005
Concentration*Wood type*Density	2884,7	2	1442,4	3,217	0,043219
Error	59175,0	132	448,3		

The ANOVA results in Table 4-8 showed a 3-way interaction between concentration, wood type and density to be statistically significant ($p<0.05$) for CCA and control wood failure percentage data.

The post-hoc (TukeyHSD) results revealed that only 4 combinations had significant differences (bars that share letters not significant, see Figure 4-12).

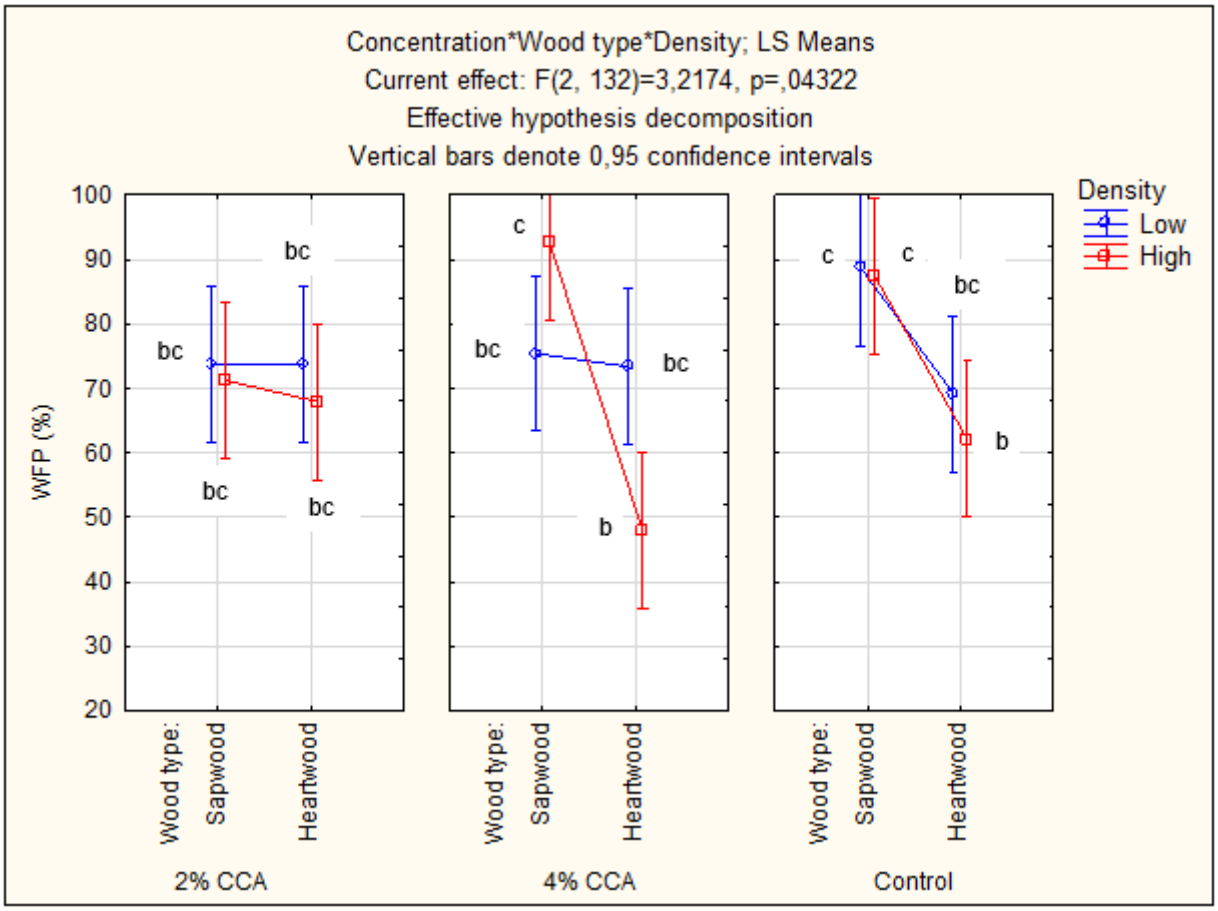


Figure 4-12: A 3-way significant interaction between density, wood type and concentration for WFP in CCA treated and untreated blocks.

The 3-way interaction in Figure 4-12 further shows that heartwood specimens with high density had a decrease in WFP. The most notable decrease in WFP was of heartwood specimens with high density, treated with 4% CCA (as shown in Figure 4-12).

The significant decrease in WFP for heartwood specimens might have been caused by the high levels of extractives present in heartwood which often interfere with bond formation. Widsten *et al.*, (2006) also found that species with poorest polyurethane gluability are characterised by relatively high bulk phenolic extractives and surface lipophilic extractives contents, as well as high density.

Secondly, the positive influence of high density on WFP, might be explained by the fact that since high density wood is usually associated with enhanced strength (contains more material per unit volume) and can carry more load, they are more likely to fail in the bond line. Whereas low density wood is generally weaker (larger lumens and thin cell walls), resulting in less stress on the bond line (Hunt *et al.*, 2019) and cause wood to fail before adhesive bond line fails. Vick (1999), also reported that although strength increased with wood density, wood failure decreases gradually up to a density range of 0.7 to 0.8 g/cm^3, and then decreases more rapidly above 0.8 g/cm^3.

Furthermore, the presence of 4% CCA preservative seemed to exacerbate the decrease of WFP in heartwood specimens. Cameron and Pizzi (1985) explained that lower wood failure in CCA treated wood could be due lack of adsorption of the adhesive onto the already heavily coated wood fibres.

Overall, these results indicate that the presence of high level of extractives in denser wood is likely to result in low wood failure.

4.3.2. DOT and control shear strength

A 3-way factorial ANOVA (3 x 2 x 2) was conducted in order to evaluate the effect of DOT preservative and certain wood properties (density and sapwood/heartwood) on the bond strength of PUR adhesive in pine laminates in comparison to untreated samples.

The shear strength data violated the assumption of normal distribution and was transformed using the Box-Cox transformation. However, the ANOVA for transformed data identified the same significant factors as the untransformed data ANOVA. As such, the ANOVA results and interaction plot displayed will be from untransformed data. The ANOVA results are presented in Table 4-9.

Table 4-9: ANOVA results for shear strength of DOT treated samples.

Effect	SS	Degr. of Freedom	MS	F	p
Intercept	15197,75	1	15197,75	11248,15	0,000000
Concentration	0,62	2	0,31	0,23	0,796177
Wood type	21,09	1	21,09	15,61	0,000126
Density	6,71	1	6,71	4,97	0,027512
Concentration*Wood type	5,93	2	2,96	2,19	0,115565
Concentration*Density	3,83	2	1,92	1,42	0,245859
Wood type*Density	0,09	1	0,09	0,07	0,792877
Concentration*Wood type*Density	21,68	2	10,84	8,02	0,000514
Error	178,35	132	1,35		

The results in Table 4-9 shows a 3-way interaction between concentration, wood type and density to be statistically significant at a 5% significance level.

The post-hoc results showed that only 5 comparison combinations had significant differences (bars that share letters not significant, see Figure 4-13)

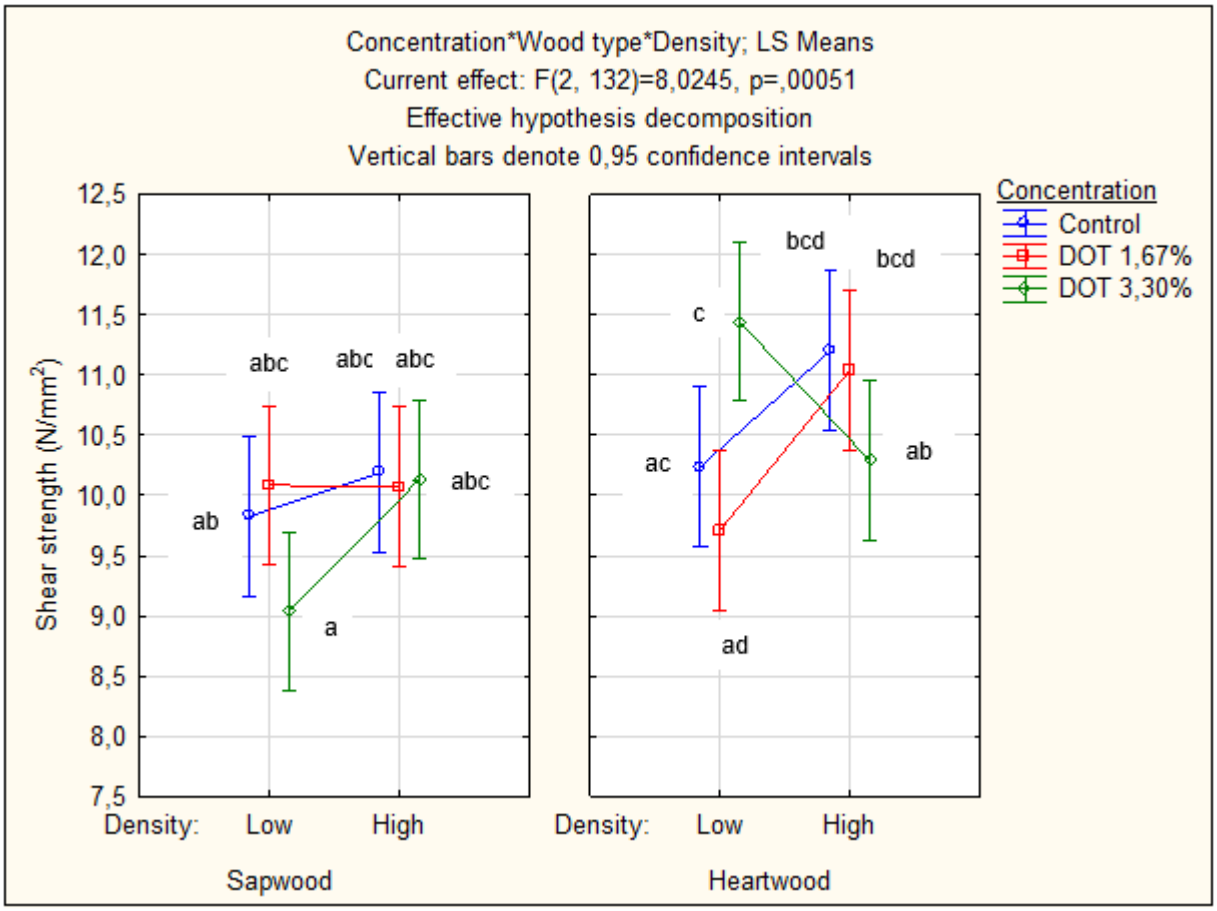


Figure 4-13: A 3-way significant interaction between concentration, wood type and density for shear strength (N/mm^2) in DOT treated and untreated blocks.

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From the interaction plot in Figure 4-13 and post-hoc results, it can be stated that the crystalline deposits from DOT preservative had no negative effect on the shear strength results. These results can be explained by the findings of Long & Morrell (2012). The authors found that the solution of DOT preservative with a concentration of 10%, did not have any significant effect on shear strength results of Douglas-fir beams bonded with resorcinol resin. In a similar study, Lesar *et al*. (2011) also found that the inclusion of boron in PUR adhesive bonded specimens increased the shear strength, however this difference was not significant. Both authors reported that crystal deposits from borate-based compounds do not interfere with bond formation.

However, the results showed a notable effect of wood properties on the shear strength. As Figure 4-13 shows, in most cases high density specimens consistently displayed a slightly higher shear strength except for heartwood specimens treated with 3.30% DOT which showed the opposite. These results suggest that the density of wood plays a significant role in the strength of bond lines and the reasons are the same as discussed with CCA in the previous section.

The reasons for the higher shear strength in heartwood samples compared to sapwood samples are the same as discussed with CCA in the previous section.

Wood failure percentage

A 3-way factorial ANOVA (3 x 2 x 2) was carried out to test the influence of DOT preservative on wood failure percentage in comparison to untreated samples. The wood failure percentage data violated the assumption of normal distribution and was transformed using the Box-Cox transformation. However, the transformed data ANOVA identified the same significant factors and interactions as the untransformed data ANOVA. As such, the ANOVA results and interaction plots displayed will be from untransformed data. The ANOVA results are presented in Table 4-10.

Table 4-10: ANOVA table for WFP of DOT treated and control blocks.

Effect	SS	Degr. of Freedom	MS	F	p
Intercept	1027689	1	1027689	4642,736	0,000000
Concentration	4739	2	2369	10,704	0,000049
Wood type	564	1	564	2,548	0,112809
Density	352	1	352	1,588	0,209802
Concentration*Wood type	6303	2	3152	14,238	0,000003
Concentration*Density	416	2	208	0,939	0,393683
Wood type*Density	0	1	0	0,001	0,977700
Concentration*Wood type*Density	444	2	222	1,003	0,369505
Error	29219	132	221		

The results in Table 4-10 found a 2-way interaction between concentration and wood type to be statistically significant at a 5% significance level for DOT and control (untreated) wood failure percentage data.

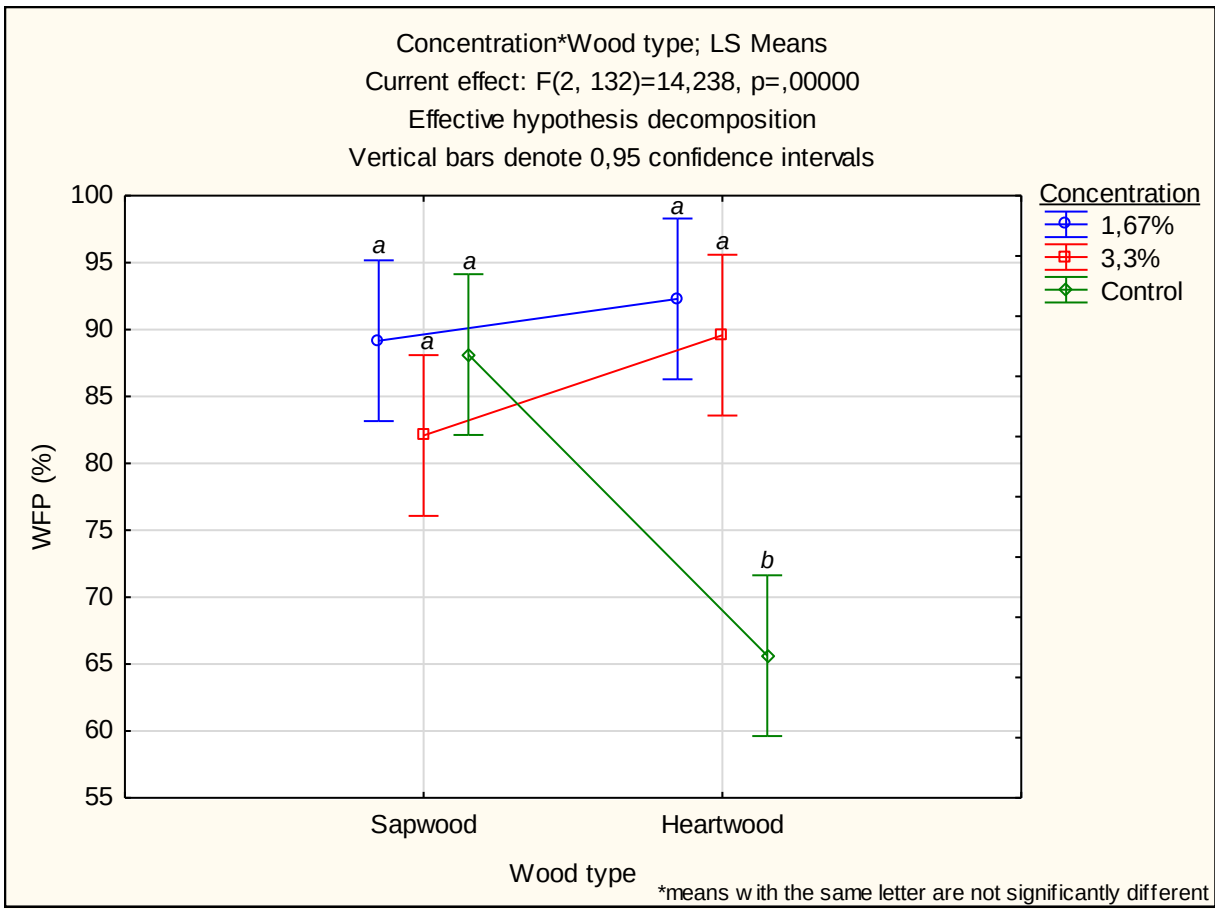

Figure 4-14: A 2-way interaction between wood type and concentration levels in DOT treated and untreated blocks for WFP.

It is evident from the results in Figure 4-14 that there was no significant difference between the wood failure percentages of DOT treated sapwood and heartwood specimens, even though the heartwood specimens showed a slightly higher wood failure. However, in the absence of DOT preservative (control), the heartwood specimens had a significantly lower wood failure percentage. Clauß *et al.* (2011) explained that the presence of extractives and aspiration of pits in heartwood can reduce the wettability of the surface and limit the penetration of adhesives and thus, lower wood failure should in expected in heartwoods.

Another interesting observation was that in the presence of DOT preservative the wood failure for heartwood specimens improved. This may indicate that the DOT crystal deposits minimized the effects caused by the heartwood extractives, which tend to interfere and obstruct with adhesion between the wood adherends and adhesive molecules. Lesar *et al*. (2011) ascribed the improved bonding quality of boron-treated wood to the crystal water in boric acid, which promotes curing.

These results suggest that the wettability of heartwood might be improved through borate-based preservatives.

4.3.3. Comparison of treatments on shear strength and wood failure

This subsection compares the shear strength results of CCA and DOT treated specimens.

In terms of EN 14080 (2013) performance requirements for individual values, all specimens treated with DOT preservative met the requirements (see Figure 4-15). The majority of CCA treated specimens also fulfilled the requirements of EN 14080 (2013), however, some of the individual specimens failed to meet the requirements (see Figure 4-15).

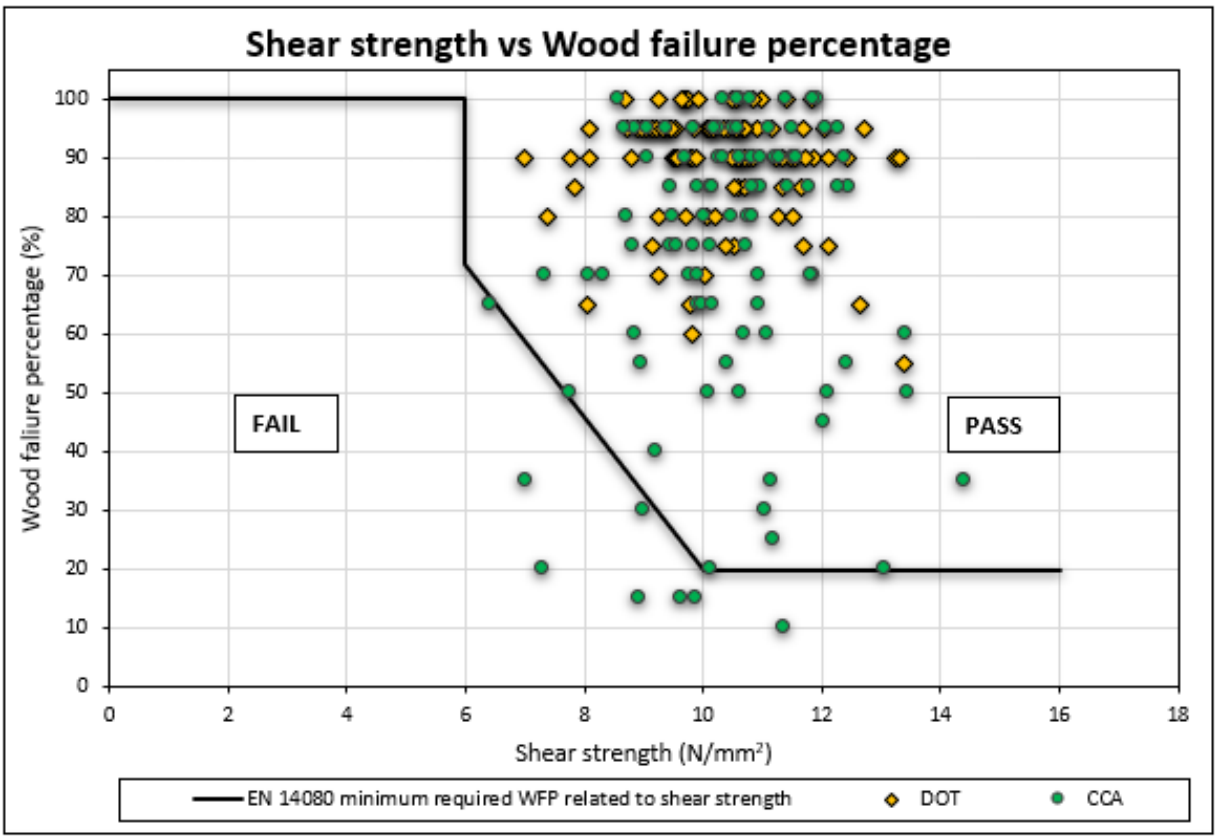

Figure 4-15: Individual block shear strength and wood failure percentage values for CCA and DOT groups in accordance with EN 14080 (2013) individual values' requirements.

In order to compare the two treatments in terms of shear strength, a one-way ANOVA was carried out. The ANOVA results were presented in a form of a mean plot (see Figure 4-16).

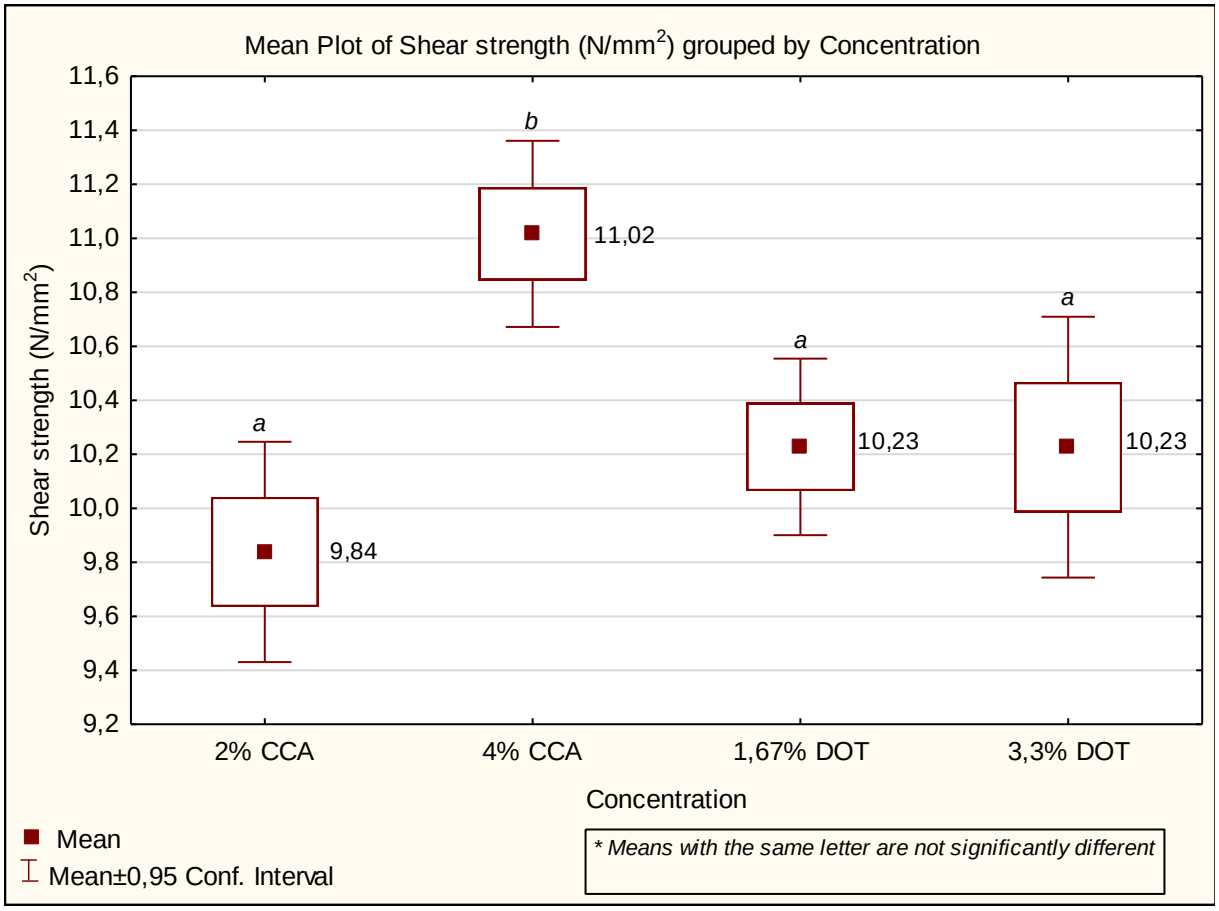

Figure 4-16: Mean shear strength for the different preservatives and treatment levels.

The 4% CCA treatment exhibited the highest shear strength (in Figure 4-16) (11.02 N/mm^2) and had a significant difference in comparison to other treatments. The DOT treatments also displayed a higher shear strength than 2% CCA, however, the difference was not significant. The high shear strength results for specimens treated with 4% CCA were unexpected, as it has been reported by Özçifçi (2006) that by increasing the CCA concentration level, the adhesion between the adhesive and wood substrate often decreases.
The higher shear strength at 4% CCA can be attributed to the fact that after treatment the chromium might be fixed to the wood surface and form a strong, stable and irreversible bond complex with the adhesive resulting in higher surface wetting by the adhesive and giving a very high strength (Cameron and Pizzi, 1985). Also, the total surface energy of CCA treated southern pine may increase with increasing retention due to the chemical modification of the wood caused by the deposition of metallic salts. The increased surface energy can improve the wettability of the surface and encourage strong adhesion between the wood adherends and adhesive molecules (Tascioglu *et al.*, 2004).

As expected, the presence of DOT showed to have no negative effect on shear strength even at increased concentration levels. Ozdemir *et al*. (2015) also reported similar findings for borate-based preservative of 1% boric acid.

In addition, the one-way ANOVA results for WFP of different preservatives and their levels is presented in Figure 4-17.

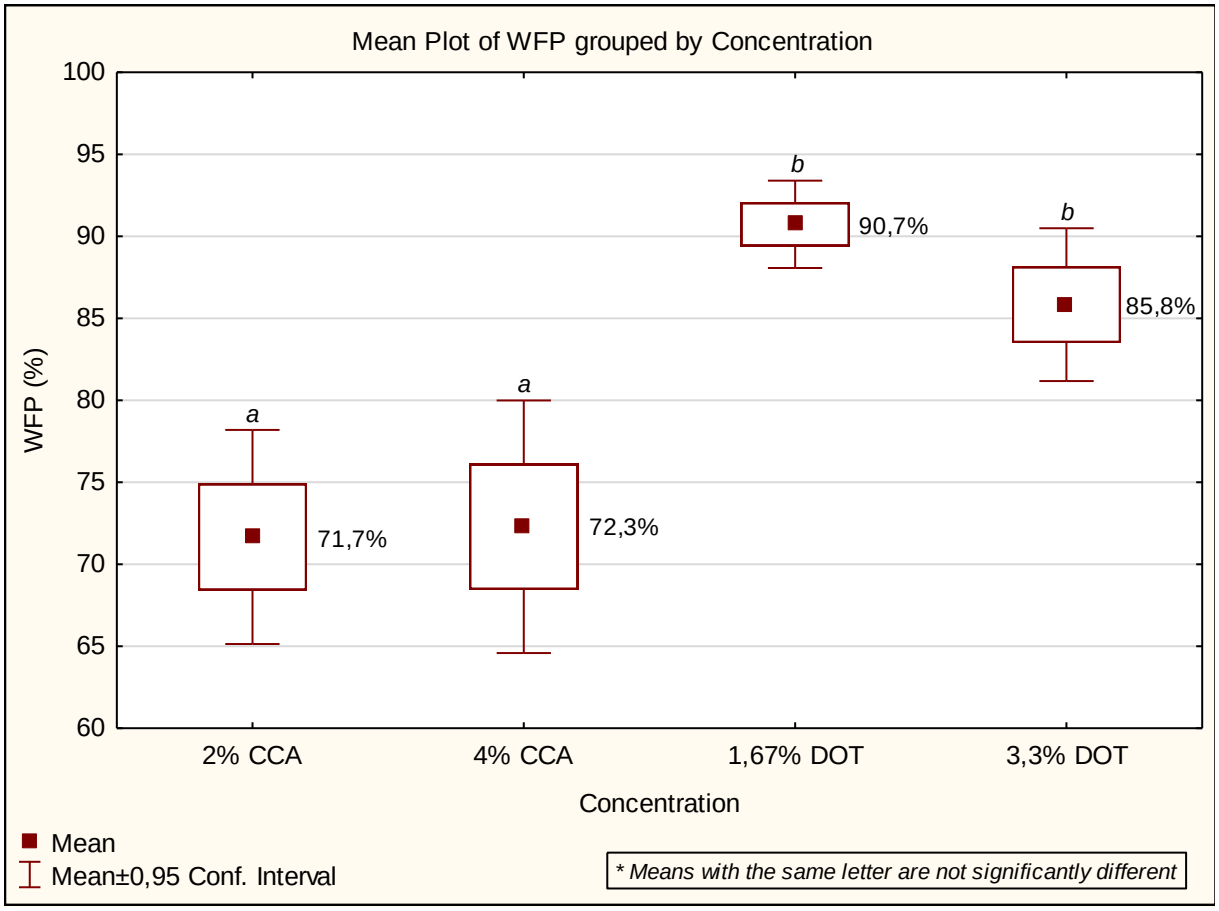

Figure 4-17: Mean WFP for different preservative and treatment levels.

The DOT treated specimens exhibited the highest wood failure percentage (see Figure 4-17) in comparison to CCA treated samples. The low wood failure percentage in CCA could have been caused by the preservative metallic deposits, which often interfere with the formation of interfacial adhesion between the wood adherents and adhesive, as they block functional sites used for anchoring by the adhesive (Cameron and Pizzi, 1985).

According to Tascioglu (2007), once the acidic CCA preservative contacts the wood, the pH increases instantly as ion-exchange and adsorption reactions occur between the metals and the wood. This leads to the CCA insoluble metal oxides occupying the functional sites of wood which are used by the adhesive for hydrogen or covalent bond formation. Cameron and Pizzi (1985) also reported that increasing CCA retention levels has a negative effect on the wood failure. This could be possibly due to lack of adsorption of the adhesive onto the already heavily coated cell lumens.

DOT treated specimens had higher wood failure compared to CCA specimens. Lesar *et al*. (2011) also reported a slight increase on the wood failure percentage on spruce specimens bonded with PUR adhesive mixed with boric acid.

Summary

This experiment aimed at evaluating the effects of waterborne preservatives (CCA and DOT) on 1C-PUR adhesive bonded laminates. Even though CCA-treated samples showed a decrease in wood failure percentage, the shear strength and wood failure percentage results still met the requirements of EN 14080 (2013). Interestingly, in some cases the CCA preservative proved to have a higher shear strength when compared to untreated samples, which was contrary to what has been reported in literature or previous findings.

Overall, the DOT-treated specimens displayed a more consistent performance in comparison to CCA specimens, in terms of shear strength and wood failure.

Furthermore, as the results demonstrated, wood properties also played a significant role in the strength of bonds. A significant positive correlation was found between density and shear strength of bond lines as specimens of high density displayed higher shear strength values. The shear strength results also showed interesting outcomes of heartwood specimens which were unexpected. According to literature the high content levels of extractives in heartwood often interfere with bond formation but based on the results of this experiment, in most cases heartwood specimens showed higher shear strength in comparison to sapwood. Such results will require further investigation in future where extractive characterisation (composition, quantity and pH of extractives, content of fatty acids etc.) is conducted, in order to identify, which extractives might have improved the bond strength in heartwood specimens of *Pinus patula*.

It should be noted that adhesive performance is dependent on the adhesive, wood species and test method and that trying to extrapolate the adhesive performance from one wood species to another is risky (Konnerth and Gindl, 2006).

4.4. Delamination: effect of CCA and DOT preservatives

The delamination test samples were prepared and tested as specified in SANS 10183-4-2, (2009), however, the test values were given a pass or fail evaluation based on *method A* of EN 14080 (2013) values displayed in Table 4-11. The reason for using a mixed approach with regards to the delamination standards, is that this research is looking to explore the preservation of engineered wood products in the South African context, hence, the use of a South African standard. However, SANS 10183-4-2 (2009) does not specify the benchmark values as EN 14080 (2013) shown in Table 4-11. It was therefore decided that test values of EN 14080 (2013) will be adopted since both standards follow a similar testing procedure.

Table 4-11: Benchmark values for delamination tests according EN 14080 (2013).

Method	**Total delamination percentage after cycle number**		
	1	**2**	**3**
A	-	5 %	10 %
B	4 %	8 %	-
C	10 %	-	-

The glueline openings (delamination) of the test blocks were measured on both end grain surfaces, after completing the three impregnating-drying cycles (*Method A*).

4.4.1. Delamination of CCA and control blocks

A 3-way factorial ANOVA (3 x 2 x 2) was conducted to evaluate the effect of CCA preservative and certain wood properties (density and sapwood/heartwood) on the bond durability of PUR adhesive in pine laminates, in comparison to untreated samples (control). The delamination data violated the assumption of normal distribution and was transformed using the Tukey's Ladder of Powers transformation. The ANOVA results are presented in Table 4-12.

Table 4-12: ANOVA table for delamination of treated and untreated blocks.

Effect	SS	Degr. of Freedom	MS	F	p
Intercept	130,6321	1	130,6321	294,7044	0,000000
Concentration	2,9184	2	1,4592	3,2920	0,042032
Wood type	2,3630	1	2,3630	5,3309	0,023407
Density	0,0785	1	0,0785	0,1771	0,674910
Concentration*Wood type	0,5072	2	0,2536	0,5722	0,566487
Concentration*Density	0,2579	2	0,1290	0,2909	0,748328
Wood type*Density	0,1608	1	0,1608	0,3628	0,548558
Concentration*Wood type*Density	1,9657	2	0,9828	2,2173	0,115241
Error	37,2342	84	0,4433		

The ANOVA analyses identified *concentration* (p = 0.04) and *wood type* (p = 0.02) as significant factors at a 5% significance level (see Table 4-12).

Concentration:

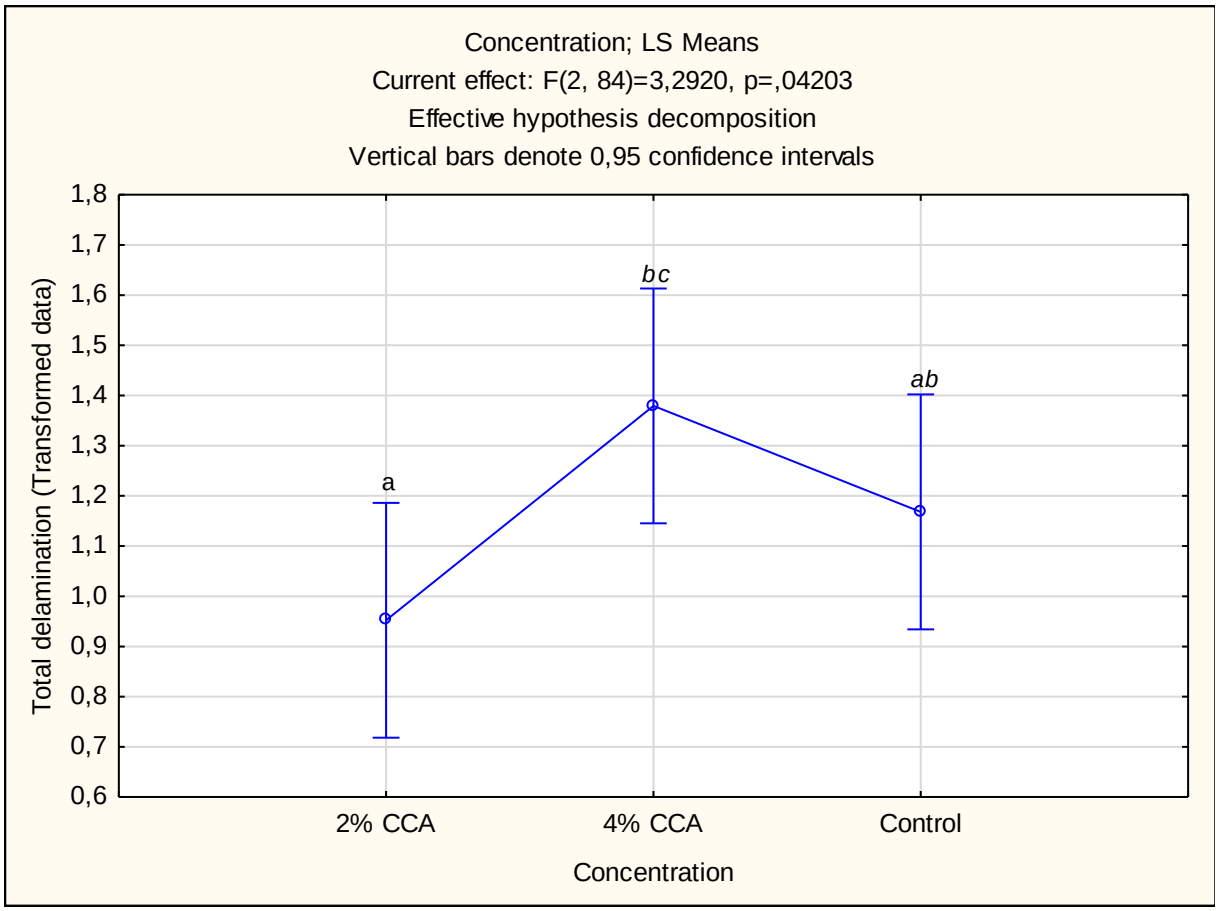

Figure 4-18: CCA and control graph for total delamination.

Figure 4-18 shows that the total delamination difference between 2% CCA and 4% CCA was statistically significant (bars that share the same letters are not significant), however, the control (untreated) test blocks had no significant difference to either of the concentration levels of CCA.

Based on the requirements of EN 14080 (2013) for total delamination (*Dtot*), all CCA-treated and untreated (control) test blocks met the requirements, as the average total delamination did not exceed 10% in length (mm) (also see Figure 4-3). These results show that despite the presence of the CCA insoluble metallic deposits which have been reported to interfere with adhesion, the adhesive bonds were able to withstand severe moisture-driven dimensional changes and produce delamination resistant bond lines at 2% and 4% CCA concentration levels. Lisperguer & Becker (2005) reported similar findings as the authors found that retention levels of CCA (4 and 6 kg/m^3) had no effect on bond durability of laboratory-synthesized PRF adhesive and met the requirements of ASTM D2559 (2004).

Hse & Kuo (1988) further highlighted that treatment with chromium-containing chemicals increases the resistance to the weathering process of wood and simultaneously fixes the water-soluble extractives.

Vick (1994) also reported that phenolic adhesives are able to produce delamination-resistant bonds in the presence of CCA metallic deposits in wood.

From the adhesive perspective, the lower delamination rate observed in the results can also be attributed to PUR adhesive, which has capabilities of absorbing additional energy upon deformation, a favourable characteristic when wood is exposed to frequent wetting and drying cycles (Lim, Tripathi and Tang, 2020).

It is particularly worth noting that although CCA-treated blocks did not exceed the maximum 10% total delamination limit, they showed an increased delamination by increasing the concentration (see Figure 4-18) and retention levels. This trend was also reported by Kang et al. (2007), where low and medium retention levels of different copper amine preservatives met the 5% ASTM D2559 (2004) requirements. However, the high retention levels failed to meet the standard requirements. Such findings indicate that careful consideration is required when bonding wood at high concentration levels/retention levels of CCA as bond durability might be affected and fail to satisfy the delamination standard requirements.

Wood type:

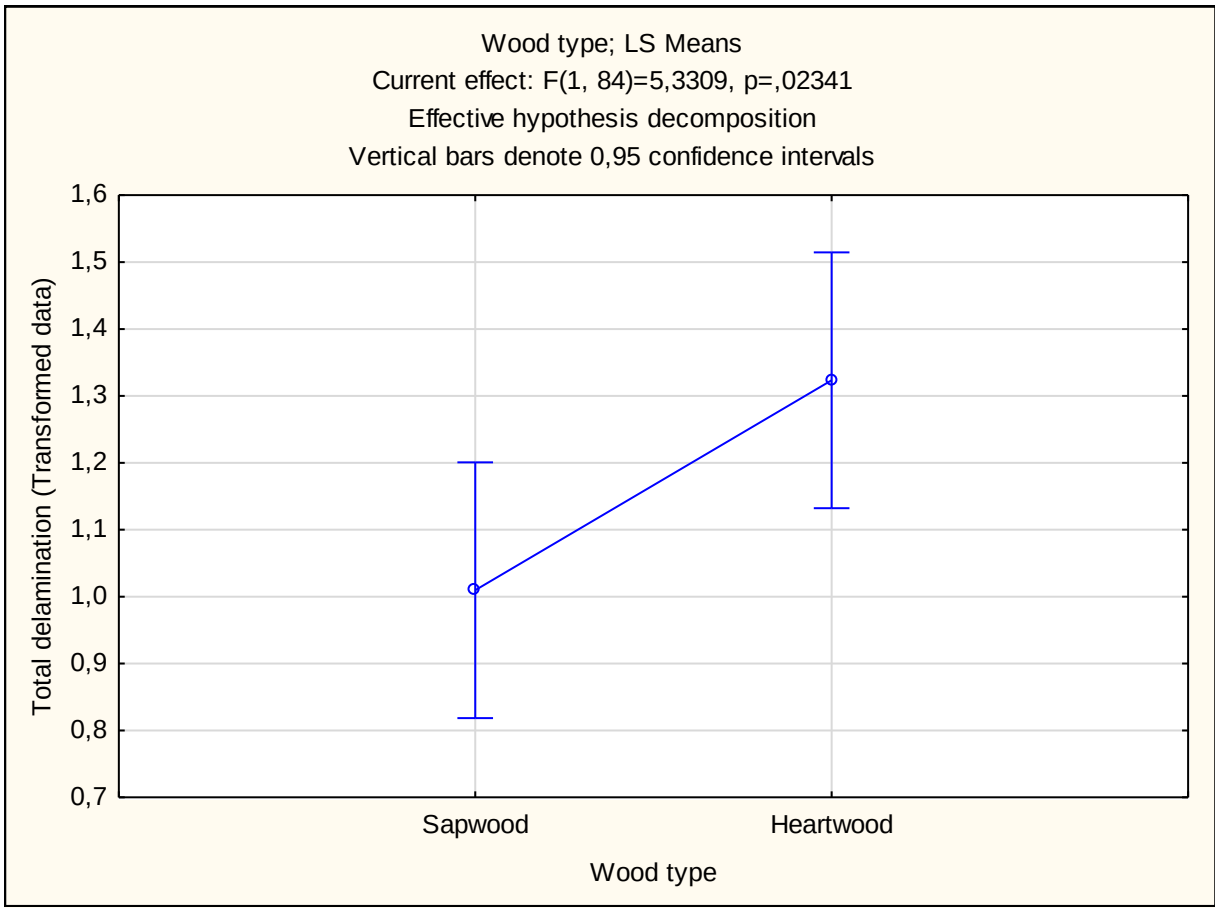

Figure 4-19: Total delamination (%) of sapwood and heartwood.

Figure 4-19 shows that the total delamination difference between sapwood and heartwood was statistically significant. Sapwood had a lower total delamination percentage.

Roffael (2016) explained that extractives of wood often have a pronounced effect on swelling and shrinkage coefficients induced by moisture change. The presence of these extractives amplifies stresses (shrinking and swelling) during the impregnating-drying cycles, resulting in larger openings on the bond line, since wood-adhesive bonds are sensitive to fast moisture content changes.

Roffael (2016) also reported that extractives can impact indirectly the strength of wood joints and the moisture-induced strain in bonded wood.

During the delamination test, it was also observed that the heartwood samples required longer drying time during the drying cycles. Knorz *et al.*, (2014) reported that the severity of the delamination test depends on the drying behaviour of the tested wood species. This may also highlight why the heartwood test blocks had a high delamination.

4.4.2. Delamination of DOT and control blocks

A 3-way factorial ANOVA (3 x 2 x 2) was conducted to evaluate the effect of DOT preservative and certain wood properties (density and sapwood/heartwood) on the bond durability of PUR adhesive in pine laminates, in comparison to untreated samples (control). The delamination data violated the assumption of normal distribution and was transformed using the Box-Cox transformation. The ANOVA results are presented in Table 4-13.

The DOT-treated and untreated (control) test blocks met the EN 14080 (2013) requirements, as the average total delamination did not exceed 10% in length (mm) (also see Figure 4-3) .

Table 4-13: ANOVA table for DOT and control delamination test blocks.

Effect	SS	Degr. of Freedom	MS	F	p
Intercept	48,40377	1	48,40377	236,1343	0,000000
Concentration	0,55858	2	0,27929	1,3625	0,261622
Wood type	0,18296	1	0,18296	0,8925	0,347499
Density	0,00284	1	0,00284	0,0138	0,906660
Concentration*Wood type	0,73250	2	0,36625	1,7867	0,173815
Concentration*Density	0,10492	2	0,05246	0,2559	0,774799
Wood type*Density	0,14059	1	0,14059	0,6859	0,409925
Concentration*Wood type*Density	0,77851	2	0,38926	1,8990	0,156097
Error	17,21866	84	0,20498		

The ANOVA results in Table 4-13 show that DOT treatment concentration or wood properties had no significant effect on delamination of PUR adhesive bond lines in *Pinus patula*.

Such outcomes were also reported in a similar experimental study conducted by Lesar et al. (2011), where it was found that PUR adhesive containing boron had no negative effect on bond durability but actually improved it. The improvement in bond durability was ascribed to crystal water in the boron acid, which promotes curing (Lesar *et al.*, 2011).

It must be pointed out that very limited literature has been published that studies the effect of borate-based preservatives on bond durability (resistance to delamination).

4.4.3. Comparison of CCA and DOT treatments on delamination

In order to compare the delamination means of CCA and DOT treated samples, a one-way analysis of variance was carried out. The ANOVA results found the means to be significantly different. The results were presented in a form of a mean plot in Figure 4-20.

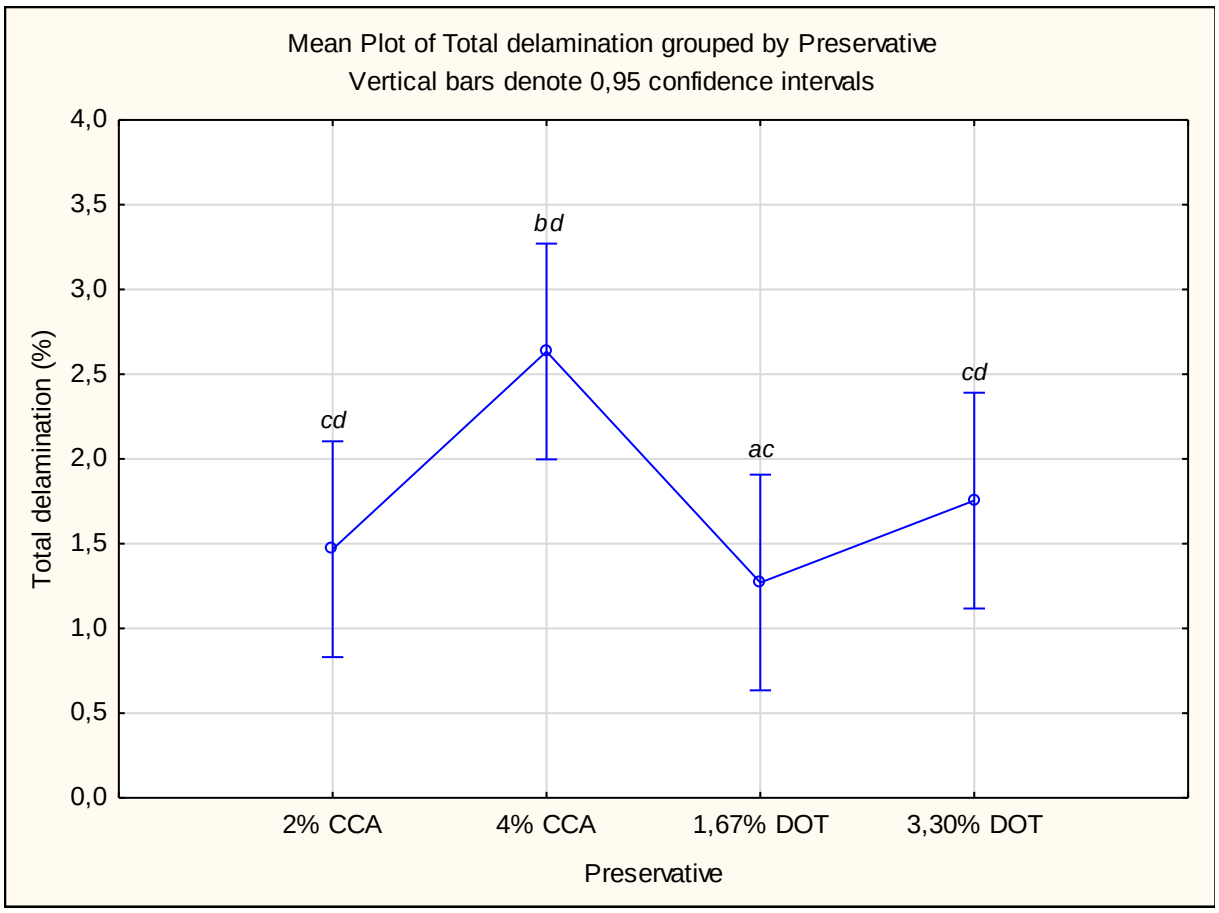

Figure 4-20: Total delamination (%) of CCA and DOT.

Figure 4-20 clearly shows that 4% CCA had the highest total delamination out of all the concentration levels tested. Such outcomes were expected as several authors have highlighted the effect CCA preservative has on bond performance. In contrast, it was interesting to note that 4% CCA also had the highest mean shear strength (see Figure 4-16).

Tascioglu (2002) concluded that laminates pre-treated with CCA negatively interfered with bond durability. Similarly, Lim et al. (2020) also found delamination to occur on bond lines of wood treated with copper-containing preservatives. This could be due to the fact that CCA metallic deposits often interfere with the intermolecular interaction between the adhesive and wood as they physically block interaction sites. Özçifçi (2006) also explained that CCA solutions having a high degree of acidity and high extent of retention, which causes the deterioration of the wood surface. Frihart & Hunt (2010) highlighted that the deposition of preservatives in wood causes air pockets and blockages which can prevent complete wetting by the adhesive and introduce stress concentration when the adhesive has cured. All these effects (caused by the CCA metallic deposits) may lead to poor adhesion between the wood substrate and adhesive and cause water to easily penetrate the bond lines and create delamination openings during bond durability testing. It is difficult however to explain why the shear strength results and bond durability results showed such contrasting results with the 4% CCA treatment.

Overall, 1.67% DOT-treated blocks had lower delamination than 4% CCA-treated blocks (see Figure 4-20). This may indicate that borate-based compounds do not significantly affect the bond performance. However, during impregnation cycles it was observed that DOT leached out of the test blocks (see Figure 4-21). This was mainly due to the high-water solubility of DOT, which is often disadvantageous. This limits the use of DOT as it cannot be used to treat wood products exposed to exterior conditions or frequent wetting or areas of high moisture content.

Figure 4-21: Leaching out of DOT in delamination test blocks.

Chapter 5 : Conclusion and Recommendations

5.1. Conclusions

Based on the results of this study, the following conclusions can be made:

Effect of wood properties: Retention rate

- In terms of retention rate, the results showed that wood properties have a significant effect on retention rate. The results showed that sapwood had a higher retention capacity than heartwood for both preservatives (CCA and DOT). Density was also found to have a significant effect on retention. In most cases, a negative relationship existed between density and retention. This indicated that the retention rate was much lower in most high-density wood samples when compared to low density-wood samples.

Bond strength – Shear strength and wood failure:

- *Pinus patula* timber treated with CCA at 2% and 4% concentration levels and bonded with 1C-PUR can be successfully used to produce engineered wood products as the shear strength and wood failure test results met the EN 14080 (2013) standard requirements. Interestingly, the 4% CCA concentration level produced the highest shear strength. However, with increasing concentration levels of CCA, the wood failure percentage decreased.
- DOT treated *Pinus patula* at 1.67% and 3.30% concentration levels, bonded with 1C-PUR can also be used to successfully produce engineered wood products as the shear strength and wood failure test results met the EN 14080 (2013) standard requirements. Overall, the DOT-treated specimens displayed more consistent performance in comparison to CCA specimens, in terms of shear strength and wood failure correlation.
- The results also indicated that wood properties play a significant role in the strength of bonds. Higher density wood had significantly better shear strength of bond lines. However, one of the unexpected findings was displayed by the heartwood test specimens. In most cases heartwood specimens showed a higher shear strength (whether treated with CCA or DOT or untreated) in comparison to sapwood.

Bond durability – Resistance to delamination:

- Despite the presence of the CCA insoluble metallic deposits, which have been reported to interfere with adhesion, the CCA adhesive bonds were able to withstand severe moisture-driven dimensional changes and meet the EN 14080 (2013) total delamination requirements and produce delamination resistant bond lines at 2% and 4% CCA concentration levels. However, it is particularly worth noting that although CCA-treated blocks did not exceed the maximum 10% total delamination limit, there was a significant difference between the low (2%) and high (4%) concentration which suggested that delamination increases with increasing concentration levels. CCA treated sapwood also displayed a lower total delamination percentage in comparison to heartwood.
- DOT-treated specimens were able to produce delamination resistant bond lines at 1.67% and 3.30% concentration levels as they met the EN 14080 (2013) standard requirements. Wood properties did not affect delamination results.

- A trend was also displayed between low and high concentration levels, for both preservatives. The low concentration levels (2% CCA and 1.67% DOT) produced lower delamination in comparison to the higher concentration levels (4% CCA and 3.33%). Such trends suggest that as the concentration level or retention rate increases, delamination is also likely to increase.

In all tests conducted (shear strength, wood failure and resistance to delamination), it can be concluded that 1C-PUR adhesive is compatible with CCA (2% and 4%) and DOT (1.67% and 3.30%) wood preservatives and can be used to produce treated engineered wood products. In general, the DOT treated samples showed more consistent results than CCA treated samples, but are limited to interior use.

5.2. Recommendations

Based on the results of this study, it is recommended that the preservative concentration levels should be carefully selected, as the results indicated that with increasing concentration levels, wood failure and delamination increase for CCA preservative. Additional research is required to investigate the surface properties of treated *Pinus patula* before bonding, by means of various analytical techniques such as environmental scanning electron microscopy (ESEM) and X-ray photoelectron spectroscopy (XPS). A surface characterization of this nature will uncover and explain the contradicting outcomes obtained in this study.

Also, further research may be required to test the effect of surface roughness, were planing is done before wood treatment.

Glossary

Charge:	wood treated in one cylinder at one time.
Wood preservation:	the process of adding adequate quantities and concentrations of toxic or repellent substances to a given wood product to upgrade its resistance to biological attack and make it highly durable
Bond strength:	the force or stress required to break a bonded assembly, with failure occurring in or near the plane of the glueline
Bond durability:	the ability of a bond to with stand ambient environment
Specimen:	the quantity taken from a sample to carry out a test
Contact angle:	measures the wettability of solid surfaces by liquid
Closed assembly time:	the time between assembly of the lamellas after glue spread and application of pressure
Interphase:	the area in which both wood cells and adhesive are present or meet
Bond line:	the homogenous area which consists only of adhesive between two adherends

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APPENDICES

APPENDIX A: Retention and shear strength results with detailed groups data

SHEAR STRENGTH SUMMARY RESULTS												
Group	Preservative	Concentration %	Wood type	Density	Average density ± SD*	Wood treatment		n	Shear test (N/mm^2)		WFP (%)	
					(kg/m^3)	Targeted retention	Retention (kg/m^3)		Mean	SD	Mean	Pass shear
1	CCA	2%	Sapwood	Low	424,6 ± 13,5	6 kg/m^3	7,71	12	9,28	0,65	74%	11/12
2	CCA	4%	Sapwood	Low	437,8 ± 20,2	12 kg/m^3	15,79	12	10,00	0,61	75%	12/12
3	CCA	2%	Sapwood	High	501,0 ± 15,8	6 kg/m^3	6,97	12	10,48	1,37	71%	11/12
4	CCA	4%	Sapwood	High	518,8 ± 8,8	12 kg/m^3	15,80	12	11,43	0,87	93%	12/12
5	CCA	2%	Heartwood	Low	435,1 ± 26,1	6 kg/m^3	8,17	12	9,34	1,29	74%	11/12
6	CCA	4%	Heartwood	Low	424,5 ± 37,2	12 kg/m^3	14,59	12	10,76	0,66	73%	10/12
7	CCA	2%	Heartwood	High	496,3 ± 29,0	6 kg/m^3	7,12	12	10,25	1,79	68%	10/12
8	CCA	4%	Heartwood	High	519,4 ± 16,7	12 kg/m^3	14,46	12	11,87	1,51	48%	11/12
9	DOT	1,67%	Sapwood	Low	449,6 ± 7,4	5 kg/m^3	7,22	12	10,09	0,74	88%	12/12
10	DOT	3,3%	Sapwood	Low	415,3 ± 11,4	10 kg/m^3	14,15	12	9,03	1,15	88%	12/12
11	DOT	1,67%	Sapwood	High	480,7 ± 11,4	5 kg/m^3	6,80	12	10,07	1,07	91%	12/12
12	DOT	3,3%	Sapwood	High	489,9 ± 13,6	10 kg/m^3	12,48	12	10,13	2,17	76%	12/12
13	DOT	1,67%	Heartwood	Low	451,8 ± 13,7	5 kg/m^3	5,64	12	9,71	0,92	93%	12/12
14	DOT	3,3%	Heartwood	Low	453,1 ± 8,5	10 kg/m^3	11,57	12	11,45	0,81	90%	12/12
15	DOT	1,67%	Heartwood	High	502,8 ± 17,1	5 kg/m^3	5,04	12	11,04	1,35	92%	12/12
16	DOT	3,3%	Heartwood	High	497,2 ± 19,8	10 kg/m^3	9,16**	12	10,29	1,38	89%	12/12
17	Control (untreated)	-	Sapwood	Low	429,4 ± 21,9	-	-	12	9,83	1,04	89%	12/12
18	Control (untreated)	-	Sapwood	High	474,8 ± 2,3	-	-	12	10,19	0,90	88%	12/12
19	Control (untreated)	-	Heartwood	Low	443,2 ± 6,6	-	-	12	10,24	0,81	69%	12/12
20	Control (untreated)	-	Heartwood	High	497,9 ± 36,2	-	-	12	11,20	0,82	62%	11/12

**Standard deviation*
***target retention not met*

APPENDIX B: Retention and delamination results with detailed groups data

TOTAL DELAMINATION SUMMARY RESULTS												
Group	Preservative	Concentration %	Wood type	Density	Average density ± SD*	Wood treatment		n	Tot. Delamination (%)			
					(kg/m^3)	Targeted retention	Retention (kg/m^3)		Mean	Min.	Max	Pass delam.
1	CCA	2%	Sapwood	Low	441,8 ± 13,4	6 kg/m^3	7,99	8	1,37%	0%	7,8%	8/8
2	CCA	4%	Sapwood	Low	439,5 ± 18,2	12 kg/m^3	14,78	8	1,61%	0,7%	3,2%	8/8
3	CCA	2%	Sapwood	High	504,6 ± 27,5	6 kg/m^3	8,01	8	1,61%	0%	3,4%	8/8
4	CCA	4%	Sapwood	High	498,6 ± 21,2	12 kg/m^3	16,49	8	1,61%	0%	4,4%	8/8
5	CCA	2%	Heartwood	Low	441,1 ± 22,2	6 kg/m^3	7,65	8	1,68%	0,2%	5,4%	8/8
6	CCA	4%	Heartwood	Low	439,4 ± 17,6	12 kg/m^3	16,30	8	2,48%	0,6%	6,3%	8/8
7	CCA	2%	Heartwood	High	510,6 ± 23,7	6 kg/m^3	5,55**	8	1,20%	0%	4,1%	8/8
8	CCA	4%	Heartwood	High	486,4 ± 13,4	12 kg/m^3	13,03	8	5,00%	1,2%	12,6%	7/8
9	DOT	1,67%	Sapwood	Low	440,7 ± 17,4	5 kg/m^3	7,67	8	0,99%	0%	6,38%	8/8
10	DOT	3,3%	Sapwood	Low	436,3 ± 13,1	10 kg/m^3	14,79	8	2,19%	0,95%	3,05%	8/8
11	DOT	1,67%	Sapwood	High	503,0 ± 19,2	5 kg/m^3	6,66	8	1,09%	0%	2,10%	8/8
12	DOT	3,3%	Sapwood	High	485,7 ± 10,0	10 kg/m^3	13,08	8	1,35%	0,38%	3,33%	8/8
13	DOT	1,67%	Heartwood	Low	444,6 ± 25,1	5 kg/m^3	5,48	8	1,66%	0,48%	3,14%	8/8
14	DOT	3,3%	Heartwood	Low	451,5 ± 24,3	10 kg/m^3	14,94	8	1,27%	0%	3,24%	8/8
15	DOT	1,67%	Heartwood	High	498,1 ± 15,4	5 kg/m^3	5,25	8	1,34%	0%	4,29%	8/8
16	DOT	3,3%	Heartwood	High	501,9 ± 25,2	10 kg/m^3	10,12	8	2,30%	0%	7,41%	8/8
17	Control (untreated)	-	Sapwood	Low	442,3 ± 16,5	-	-	8	1,65%	0%	7,1%	8/8
18	Control (untreated)	-	Sapwood	High	475,9 ± 2,9	-	-	8	1,97%	0%	5,2%	8/8
19	Control (untreated)	-	Heartwood	Low	443,5 ± 15,8	-	-	8	3,22%	0,3%	11,1%	7/8
20	Control (untreated)	-	Heartwood	High	497,6 ± 25,2	-	-	8	1,73%	0%	3,2%	8/8

**Standard deviation*

***target retention not met*